Fractional spin dynamics in the Kitaev model under a magnetic field

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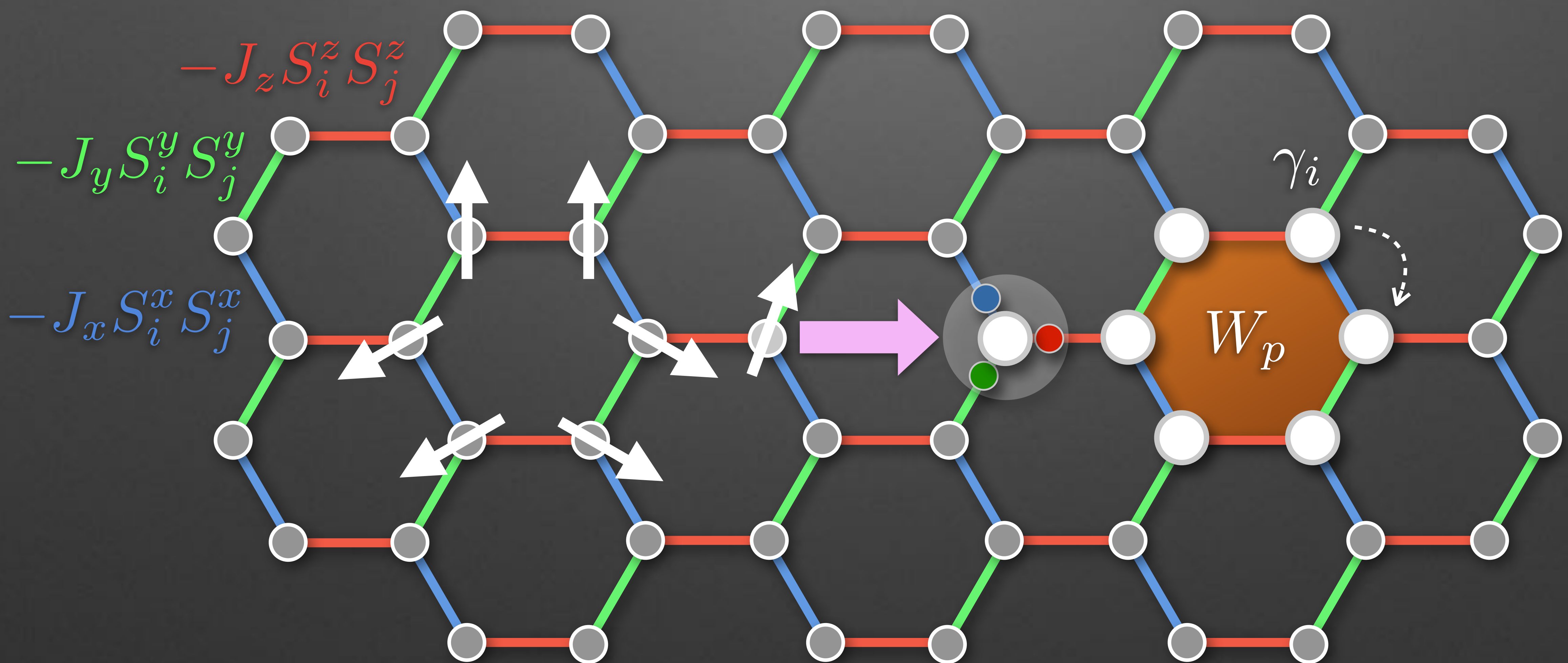
  - $4f^1$ electron compounds A_2PrO_3
  - polar spin-orbit Mott insulators

Kitaev model

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

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



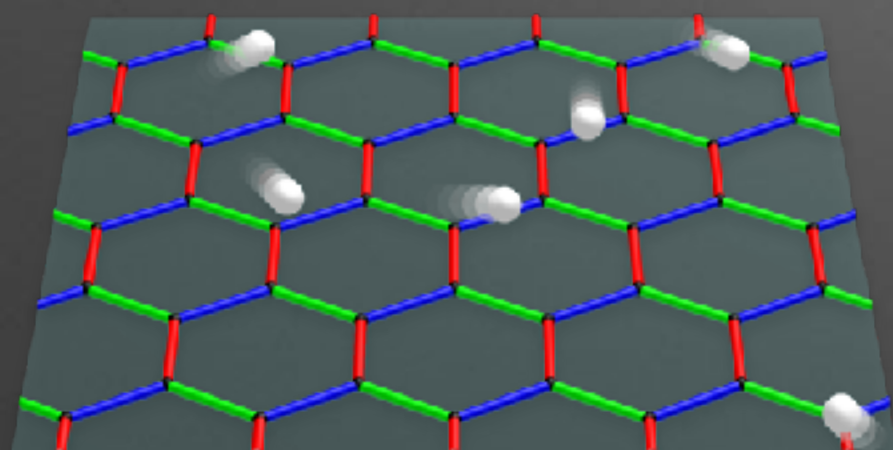
strongly frustrated, but exactly solvable
→ quantum spin liquid in the ground state

Spins are fractionalized into
itinerant Majorana fermions
and localized Z_2 fluxes

Fractional excitations

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

ground state

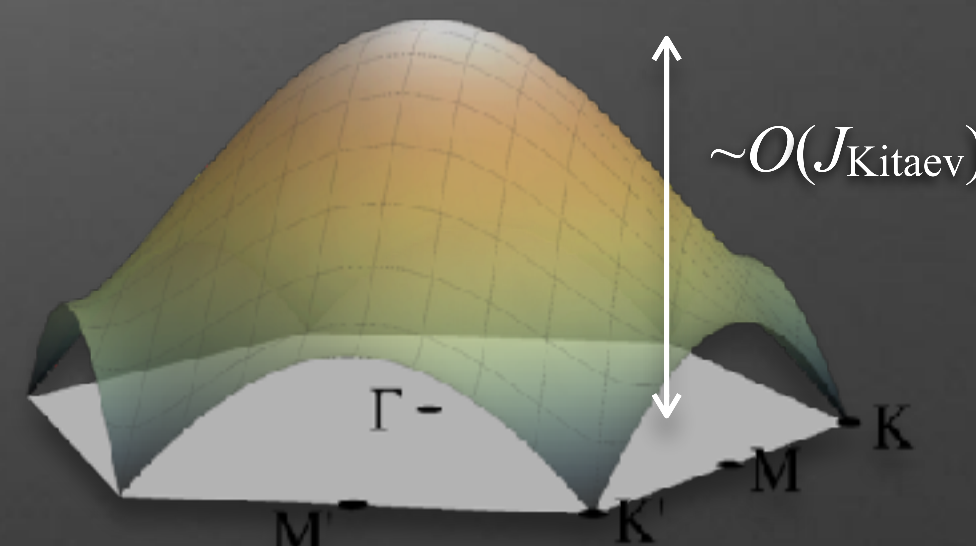


itinerant Majorana fermions on flux-free background
(all W_p=+1)

formally similar to Dirac electrons on the honeycomb lattice

excitations

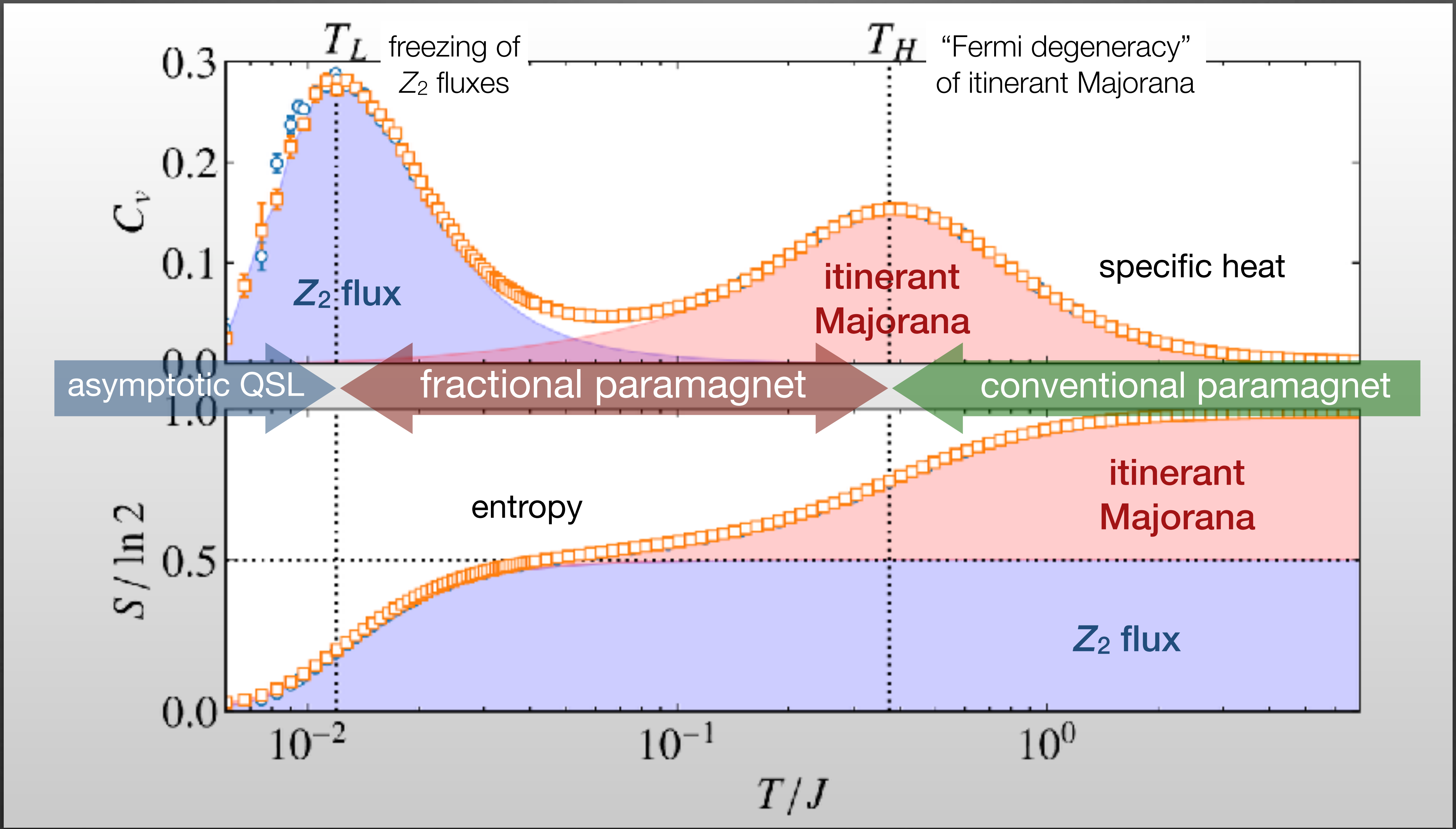
- Dirac-like dispersion for itinerant Majorana fermion excitations



- localized flux excitation (W_p=+1→-1) is always gapped and **q** independent

$$\Delta_{\mathrm{flux}} \sim 0.06\ J_{\mathrm{Kitaev}}$$

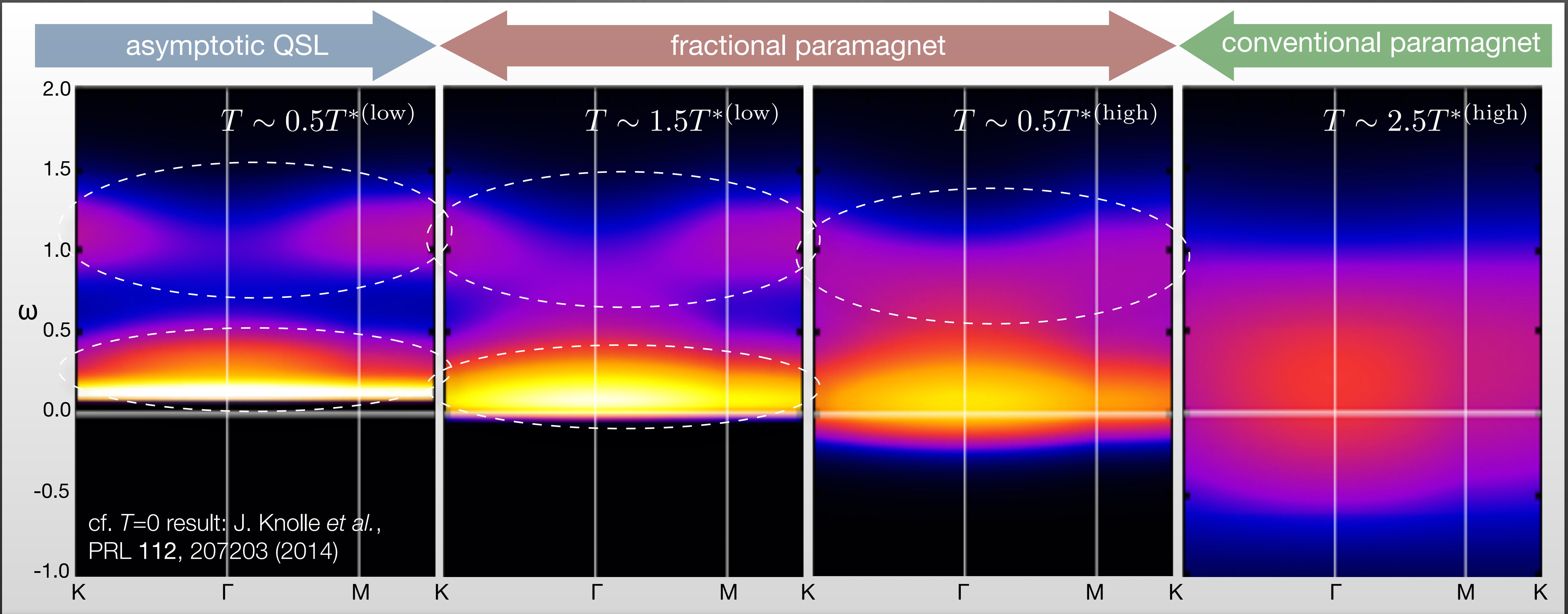
Thermal fractionalization



J. Nasu, M. Udagawa, and YM, PRB **92**, 115122 (2015)
for review, see YM and J. Nasu, preprint (arXiv:1909.02234)

Fractional spin dynamics

dynamical spin structure factor $S(\mathbf{q},\omega)$

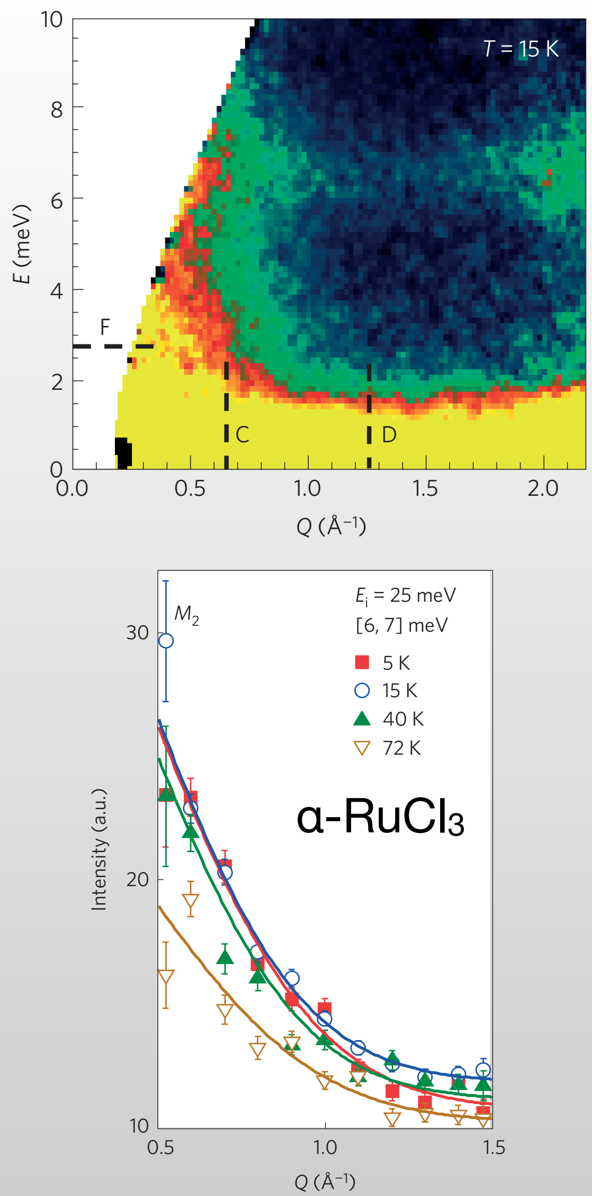


dichotomy of spin excitation:

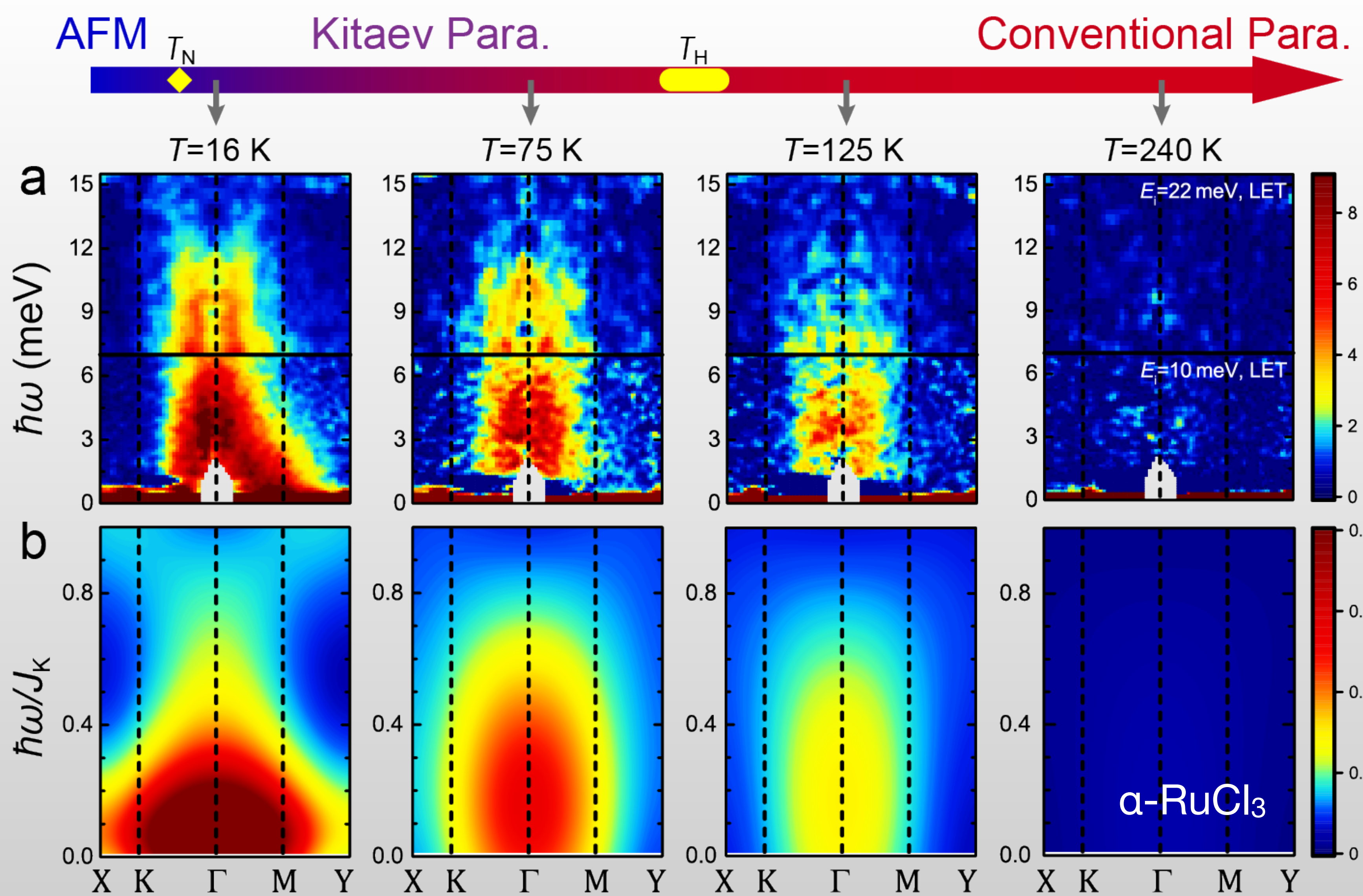
- growth of high-energy continuum at $\omega \sim J$ in the fractional PM region
- growth of quasi-elastic response toward the asymptotic QSL region

J. Yoshitake, J. Nasu, and YM, Phys. Rev. Lett. **117**, 157203 (2016)
J. Yoshitake, J. Nasu, Y. Kato, and YM, Phys. Rev. B **96**, 024438 (2017)
J. Yoshitake, J. Nasu, and YM, Phys. Rev. B **96**, 064433 (2017)

Comparison with experiment



A. Banerjee *et al.*, Nat. Mater. **15**, 733 (2016)



S.-H. Do, S.-Y. Park, J. Yoshitake, J. Nasu, YM *et al.*, Nat. Phys. **13**, 1709 (2017)

good agreement in T,**q**,ω dependence ➡ strong signature of fractional excitations

Question

- Fractional excitations in the Kitaev QSL have been identified in both theory and experiment <u>at zero magnetic field</u>.
- Recently, many interesting aspects have been revealed in experiments <u>in an applied magnetic field</u>.
  - collapse of antiferromagnetic order and field-induced spin liquid state
  - half-quantization of the thermal conductivity, etc.

What happens to the Kitaev QSL and fractional excitations in an applied magnetic field?

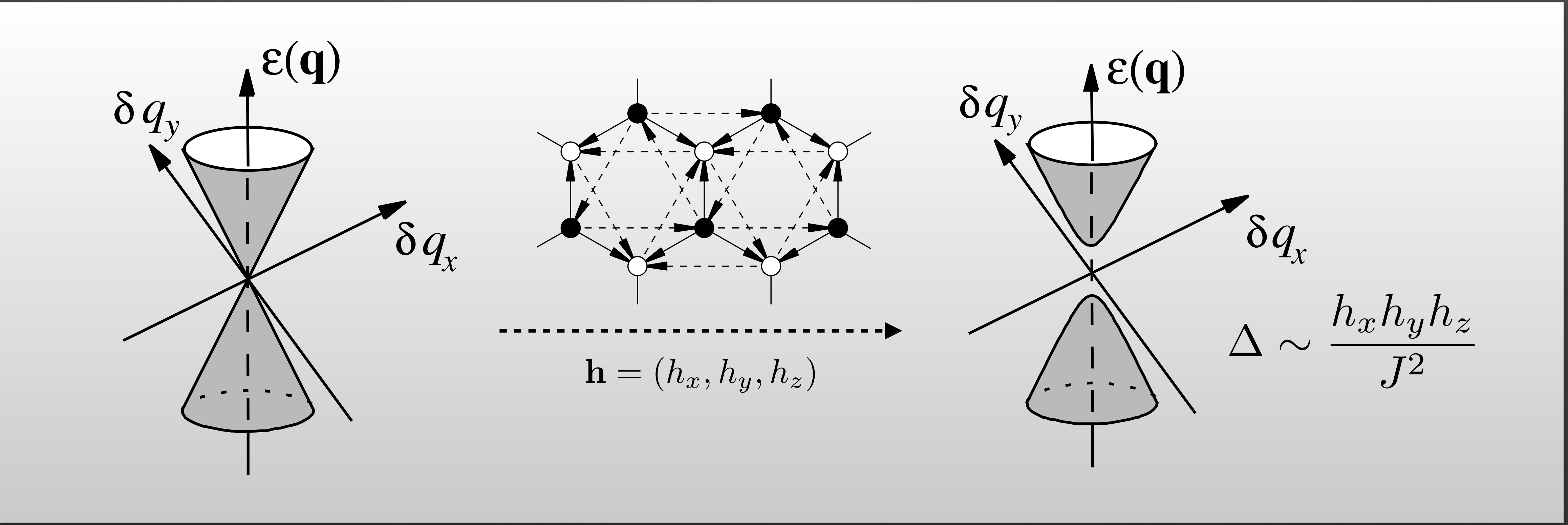
even theoretically challenging since the exact solvability is lost in the field

Perturbation theory

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

- lowest-order contribution from the Zeeman coupling to $\boldsymbol{h}=(h_x, h_y, h_z)$

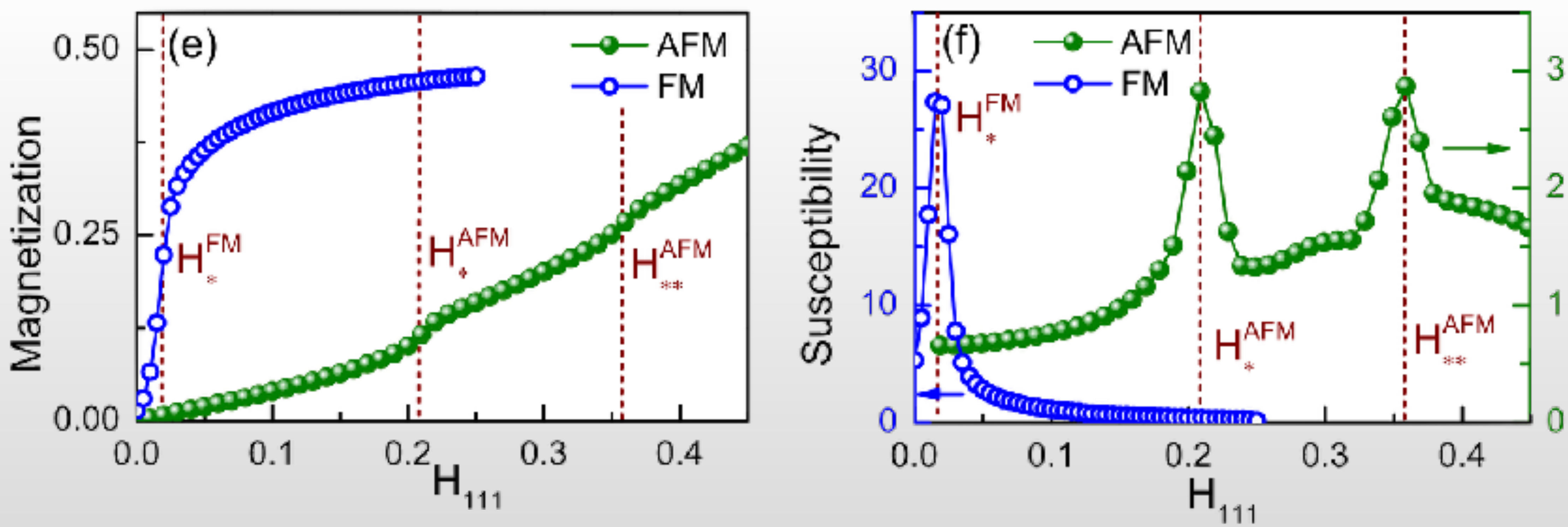
$$\mathcal{H}^{\text{eff}} \sim -\frac{h_x h_y h_z}{J^2} \sum_{j,k,l} \sigma_j^x \sigma_k^y \sigma_l^z$$ ➡ 2nd-neighbor hopping of Majorana fermions



Magnetic field opens a mass gap in the Majorana cones:
"Chern insulator" with a chiral Majorana edge mode

Beyond perturbation

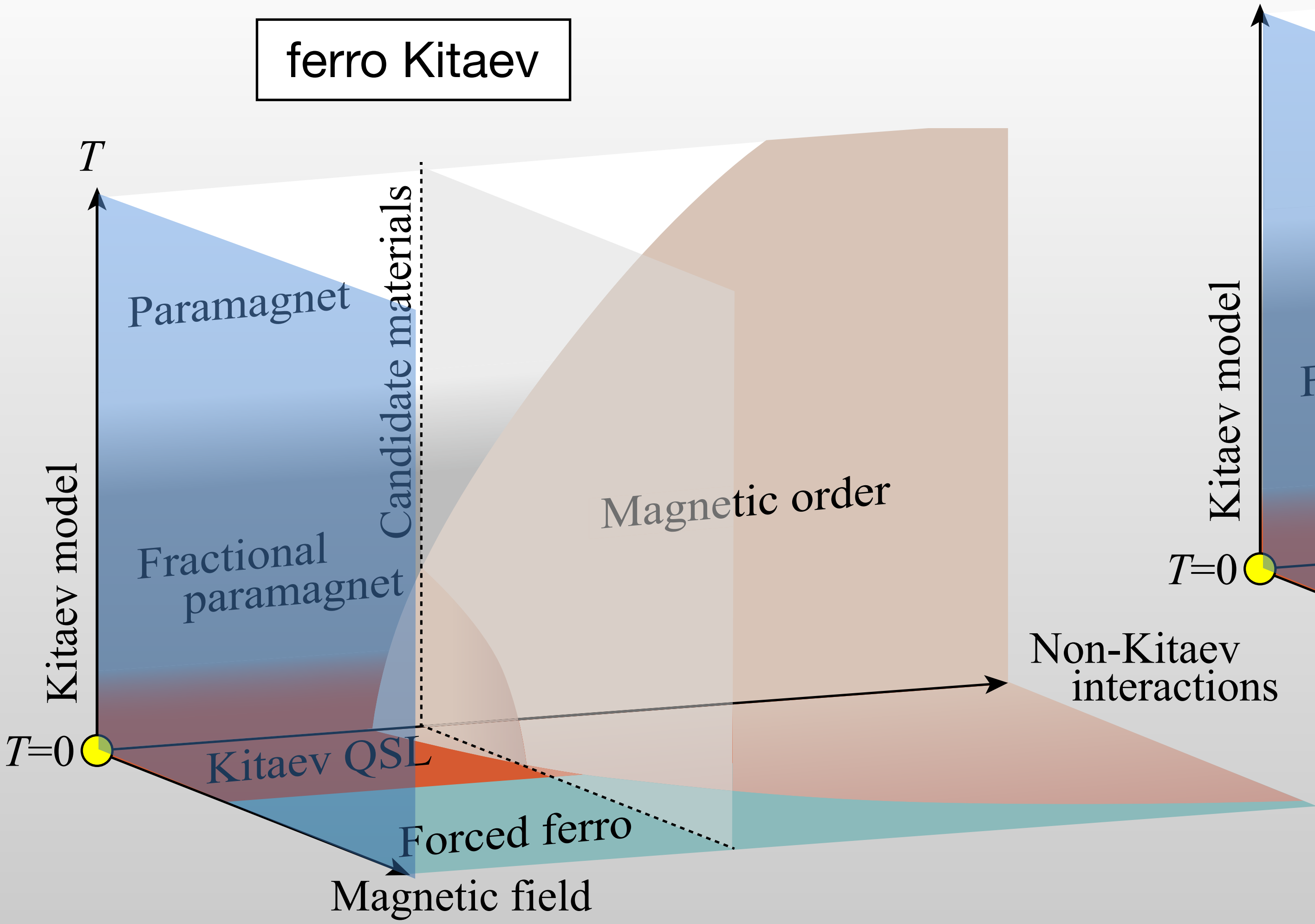
- The topological QSL survives up to nonzero field, but different behaviors appear between the ferro and antiferro Kitaev models.

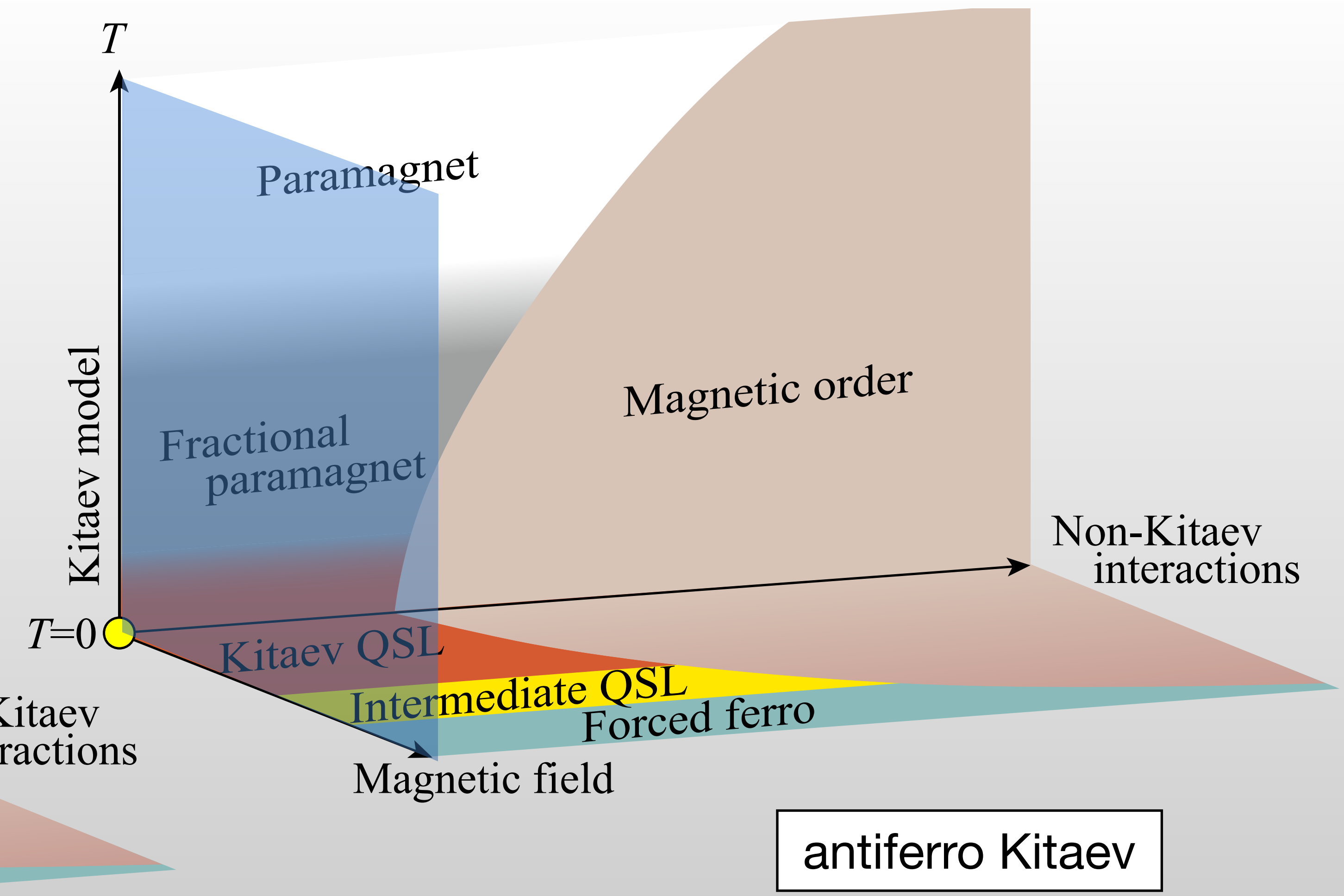


ferro case: H.-C. Jiang, Z.-C. Gu, X.-L. Qi, and S. Trebst, Phys. Rev. B **83**, 245104 (2011), …

antiferro case: Z. Zhu, I. Kimchi, D. N. Sheng, and L. Fu, Phys. Rev. B **97**, 241110(R) (2018)
M. Gohlke, R. Moessner, and F. Pollmann, Phys. Rev. B **98**, 014418 (2018)
J. Nasu, Y. Kato, Y. Kamiya, and YM, Phys. Rev. B **98**, 060416(R) (2018)
S. Liang, M.-H. Jiang, W. Chen, J.-X. Li, and Q.-H. Wang, Phys. Rev. B **98**, 054433 (2018)
C. Hickey and S. Trebst, Nat. Commun. **10**, 530 (2019)
D. C. Ronquillo, A. Vengal, and N. Trivedi, Phys. Rev. B **99**, 140413(R) (2019), …

Anticipated phase diagram





How do these phase diagrams really look like?

➡ We study the pure Kitaev case as the first step.

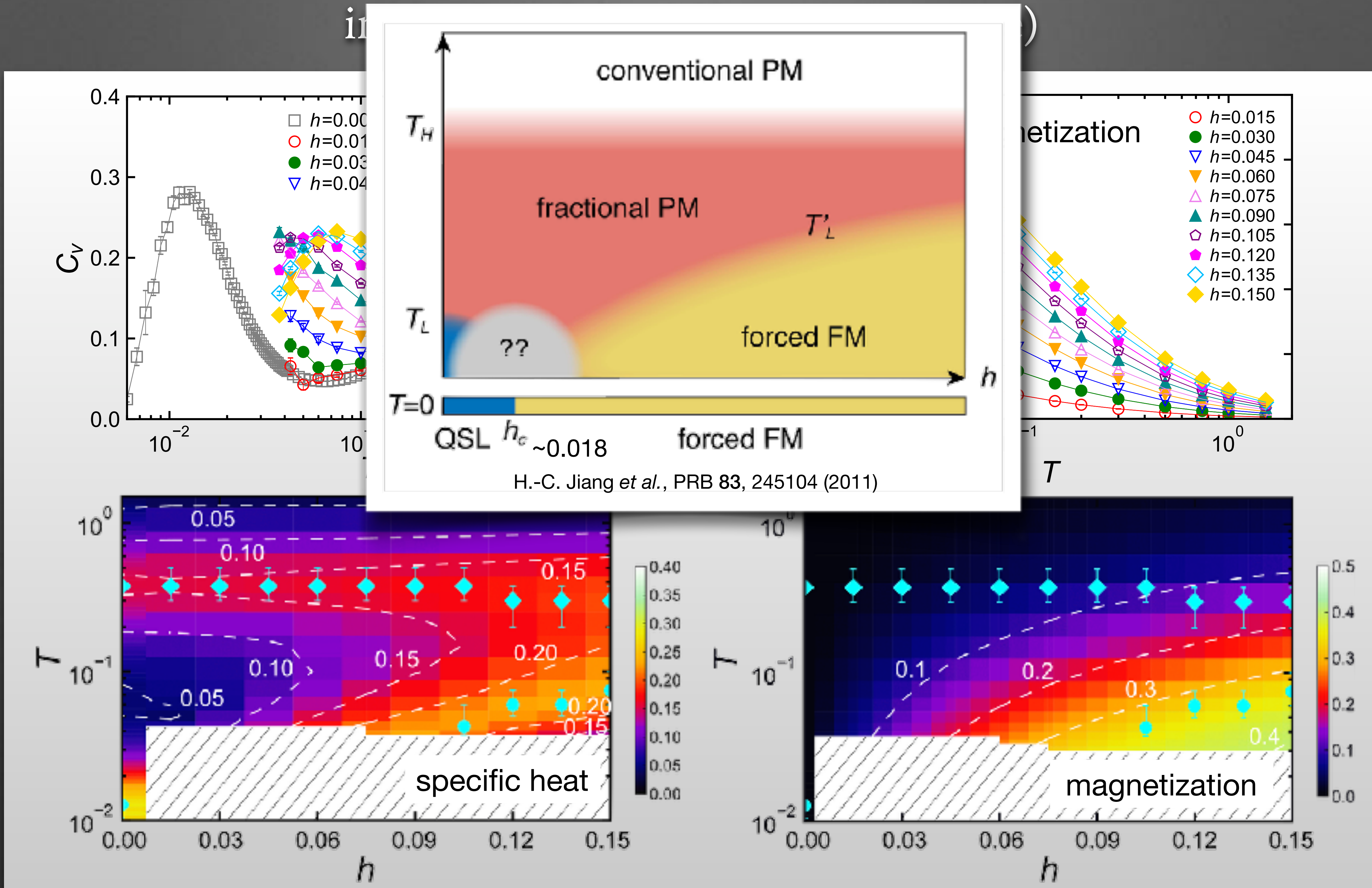
Numerical methods

- for finite T at zero field
  - Majorana-based quantum Monte Carlo
    J. Nasu, M. Udagawa, and YM, PRL **113**, 197205 (2014); P. A. Mishchenko, Y. Kato, and YM, PRB **96**, 125124 (2017)
  - Majorana-based cluster DMFT + continuous-time QMC (CTQMC)
    J. Yoshitake, J. Nasu, and YM, PRL **117**, 157203 (2016); J. Yoshitake, J. Nasu, Y. Kato, and YM, PRB **96**, 024438 (2017)
  - Majorana-QMC + CTQMC J. Yoshitake, J. Nasu, and Y. Motome, PRB **96**, 064433 (2017)
- for finite T at nonzero field
  - NEW spin-cluster-based CTQMC J. Yoshitake, J. Nasu, Y. Kato, and YM, arXiv:1907.07299

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

✓ **systematic analysis of finite-size effects:**
applicable to large enough clusters up to several 10^2 sites

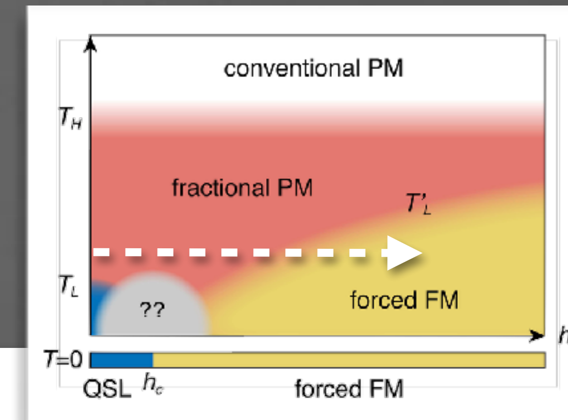
Magnetization and specific heat



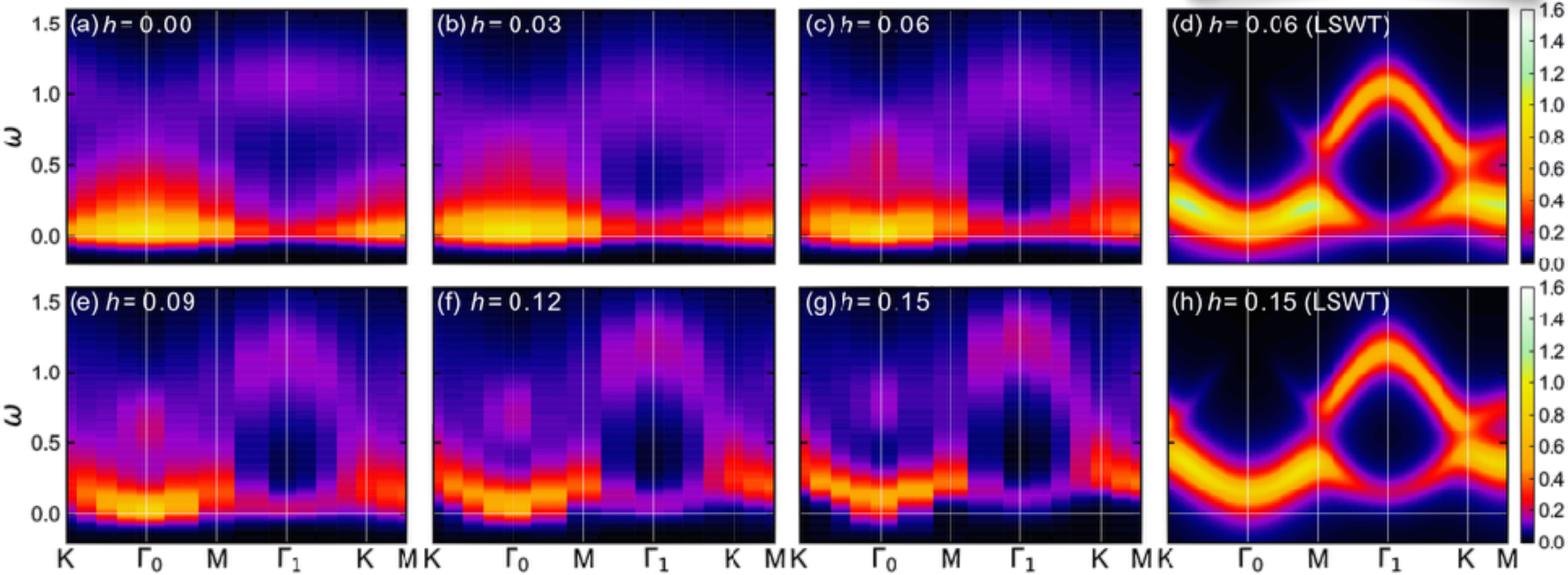
J. Yoshitake, J. Nasu, Y. Kato, and YM, preprint (arXiv:1907.07299)

cf. experimental data for the specific heat in the field: S. Widmann *et al.*, Phys. Rev. B 99, 094415 (2019)

$S(\mathbf{q},\omega)$
in the [111] field



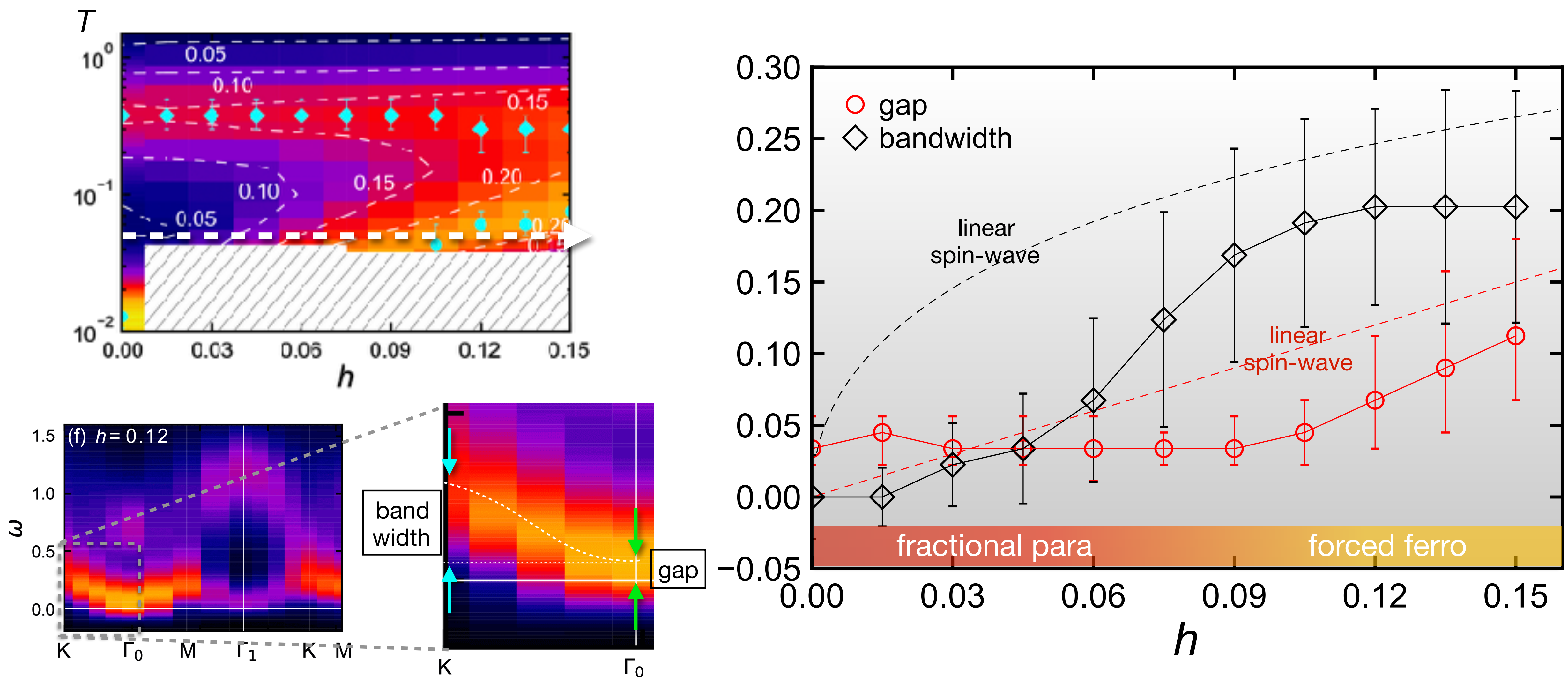
$T_L \simeq 0.012 < T = 0.05 < T_H \simeq 0.375$



- almost unchanged up to h~$0.06J$, where the Kitaev QSL is retained at T=0
- spin-wave like dispersions develop gradually above h~$0.06J$
- crossover from fractional Majorana to magnon: confinement-deconfinement

J. Yoshitake, J. Nasu, Y. Kato, and YM, preprint (arXiv:1907.07299)

Majorana-magnon crossover

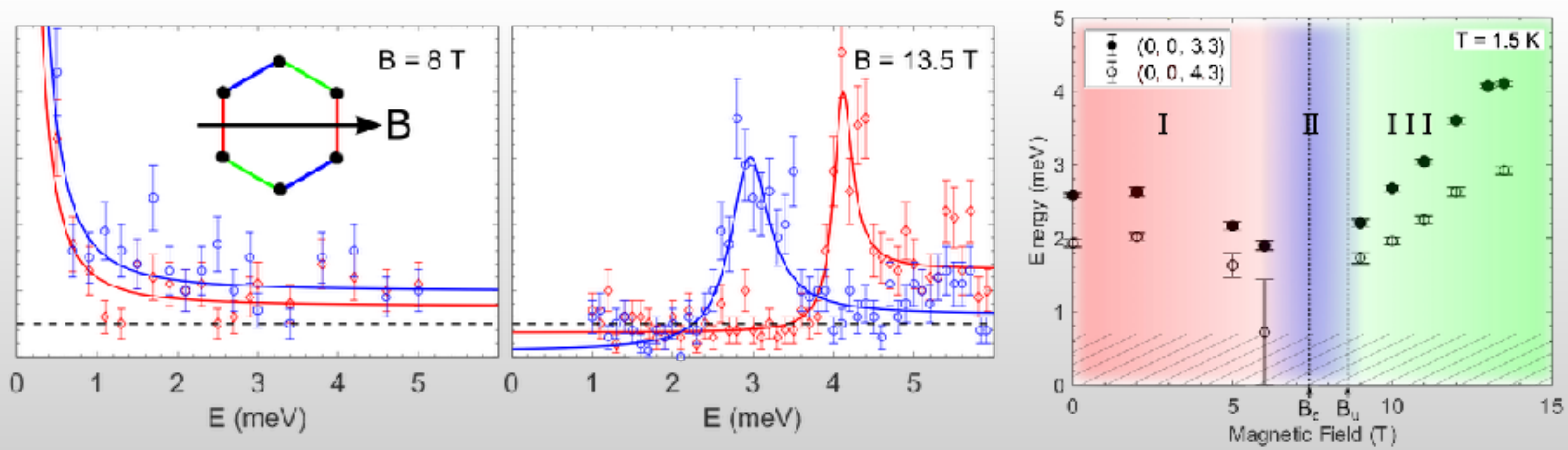


almost unchanged in the fractional paramagnetic region, but rapidly approaching the linear spin-wave dispersion in the forced ferromagnetic region

J. Yoshitake, J. Nasu, Y. Kato, and YM, preprint (arXiv:1907.07299)

Inelastic neutron scattering

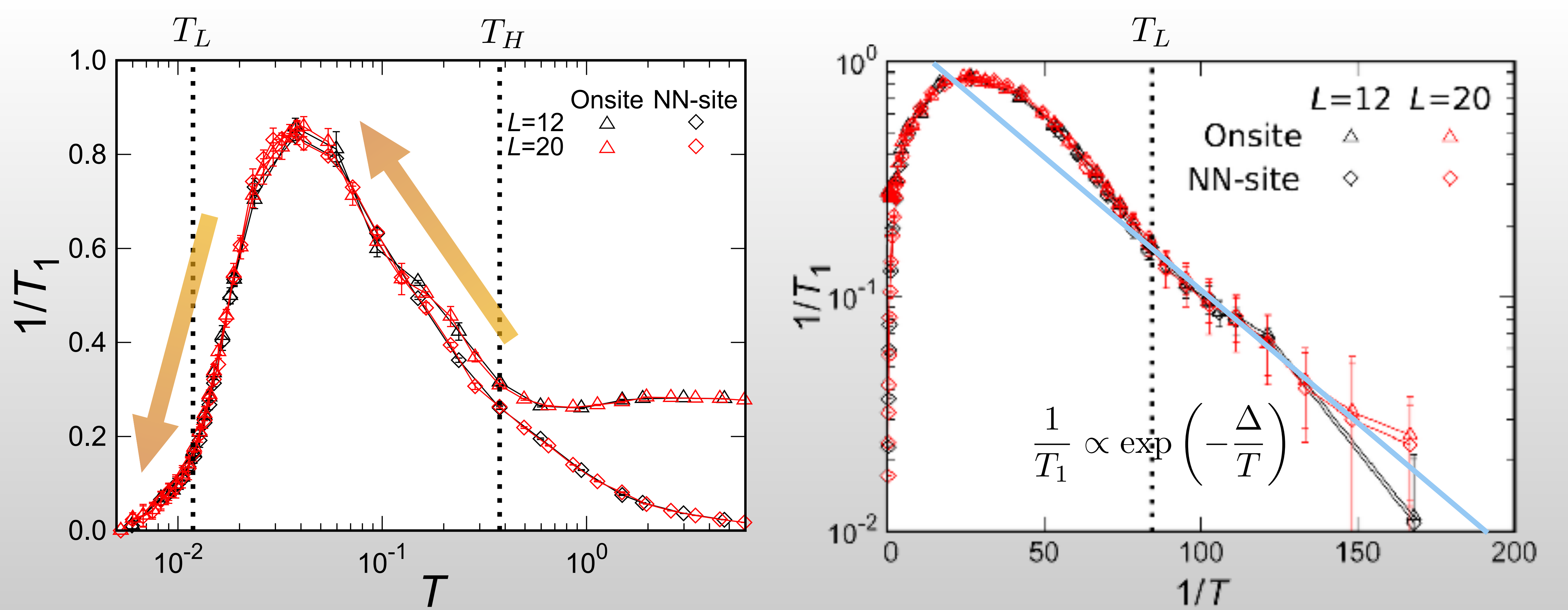
- incoherent spin excitation in the field-induced paramagnetic state, dispersive magnon-like excitation in the higher-field region



C. Balz *et al.*, Phys. Rev. B **100**, 060405(R) (2019)

NMR relaxation rate $1/T_1$

at $h=0$



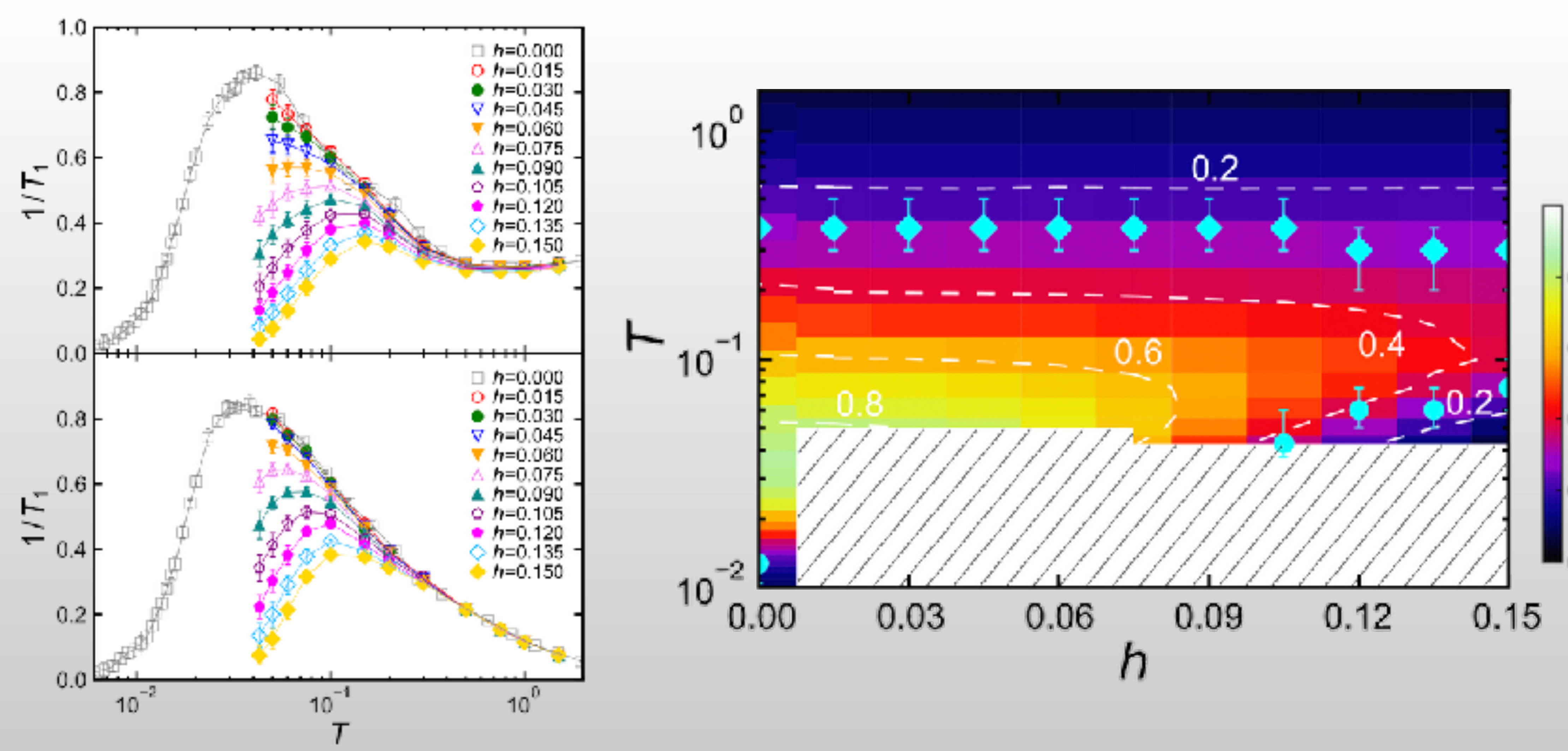
- increase below T_H down to slightly above T_L
- exponential suppression around and below T_L: flux gap opening

➡ **distinct behavior from static spin correlations: dichotomy**

J. Yoshitake, J. Nasu, and YM, Phys. Rev. Lett. 117, 157203 (2016)
J. Yoshitake, J. Nasu, Y. Kato, and YM, Phys. Rev. B 96, 024438 (2017)
J. Yoshitake, J. Nasu, and YM, Phys. Rev. B 96, 064433 (2017)

NMR relaxation rate $1/T_1$

in [111] field

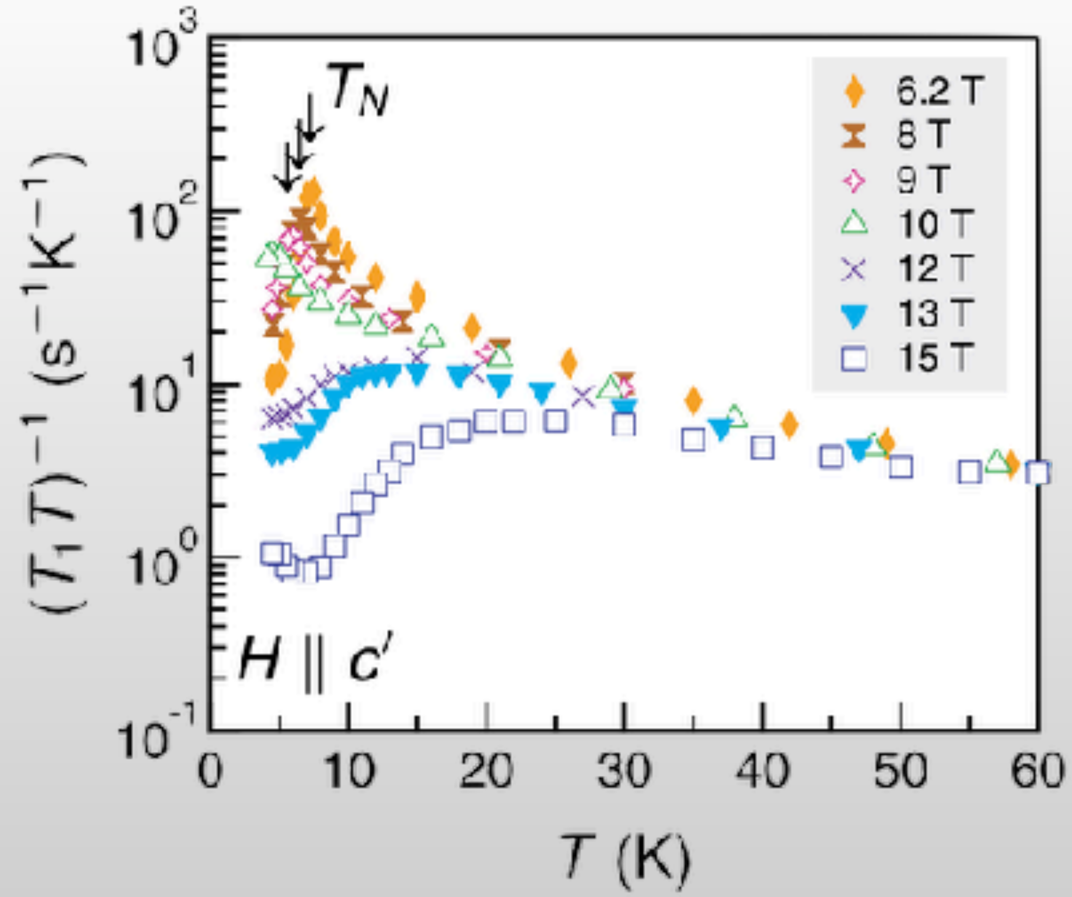


While the peak is reduced and shifted to high T, the enhancement of $1/T_1$ remains in the fractional paramagnetic region in the field.

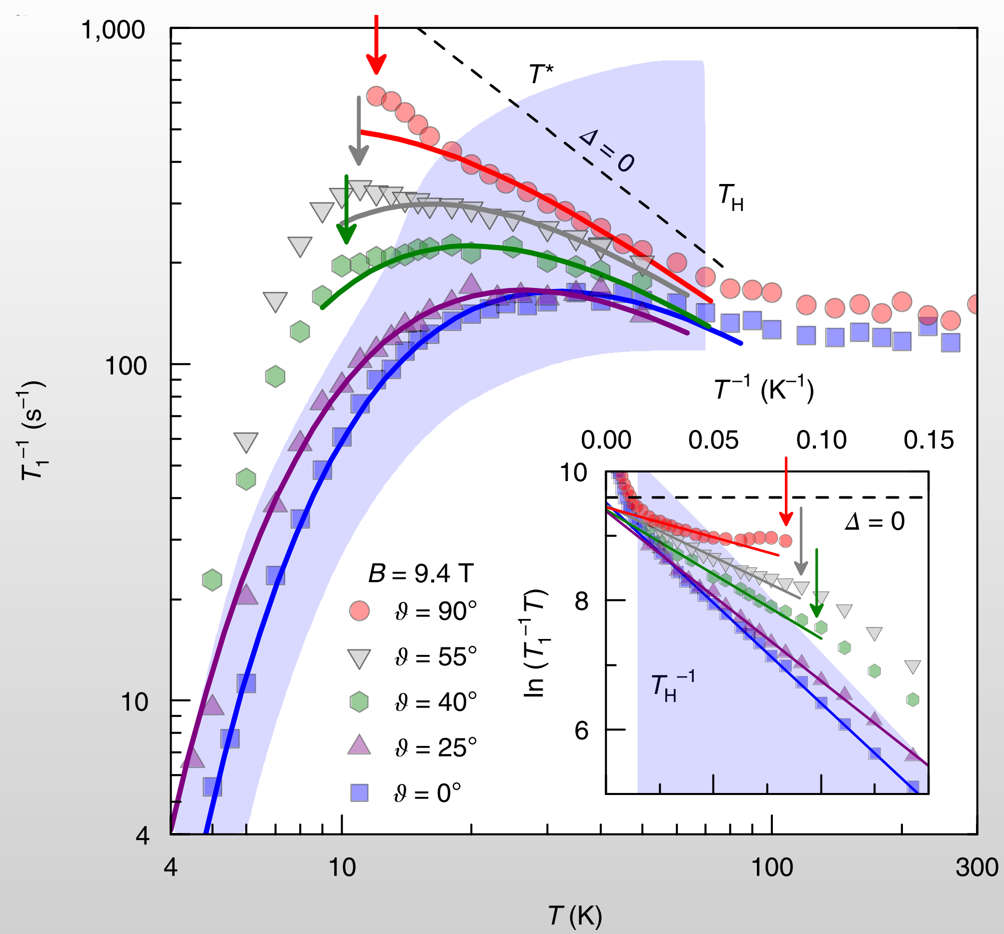
J. Yoshitake, J. Nasu, Y. Kato, and YM, preprint (arXiv:1907.07299)

NMR $1/T_1$: experiment

- good agreement with our theory in the field-induced quantum disordered region where the antiferromagnetic order is suppressed



S.-H. Baek *et al.*, Phys. Rev. Lett. **119**, 037201 (2017)



N. Jansa *et al.*, Nat. Phys. **14**, 786 (2018)

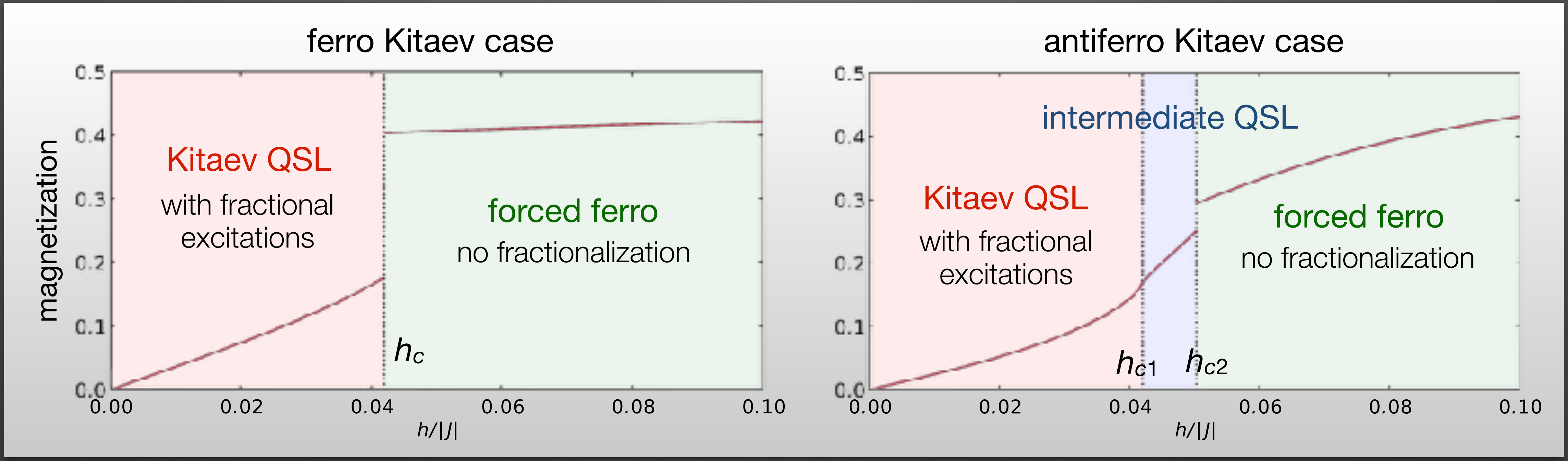
NB. further lower-T data are available in Y. Nagai, T. Jinno, Y. Yoshitake, J. Nasu, YM, M. Itoh, and Y. Shimizu, preprint (arXiv:1810.05379)

Real-time dynamics of fractional excitations by field quench

Majorana MF theory

based on the Jordan-Wigner transformation

- well reproduce the ground-state phase diagrams in the [001] field



J. Nasu, Y. Kato, Y. Kamiya, and YM, Phys. Rev. B 98, 060416(R) (2018)
cf. m-VMC study: K. Ido and T. Misawa, preprint (arXiv:1906.07325)

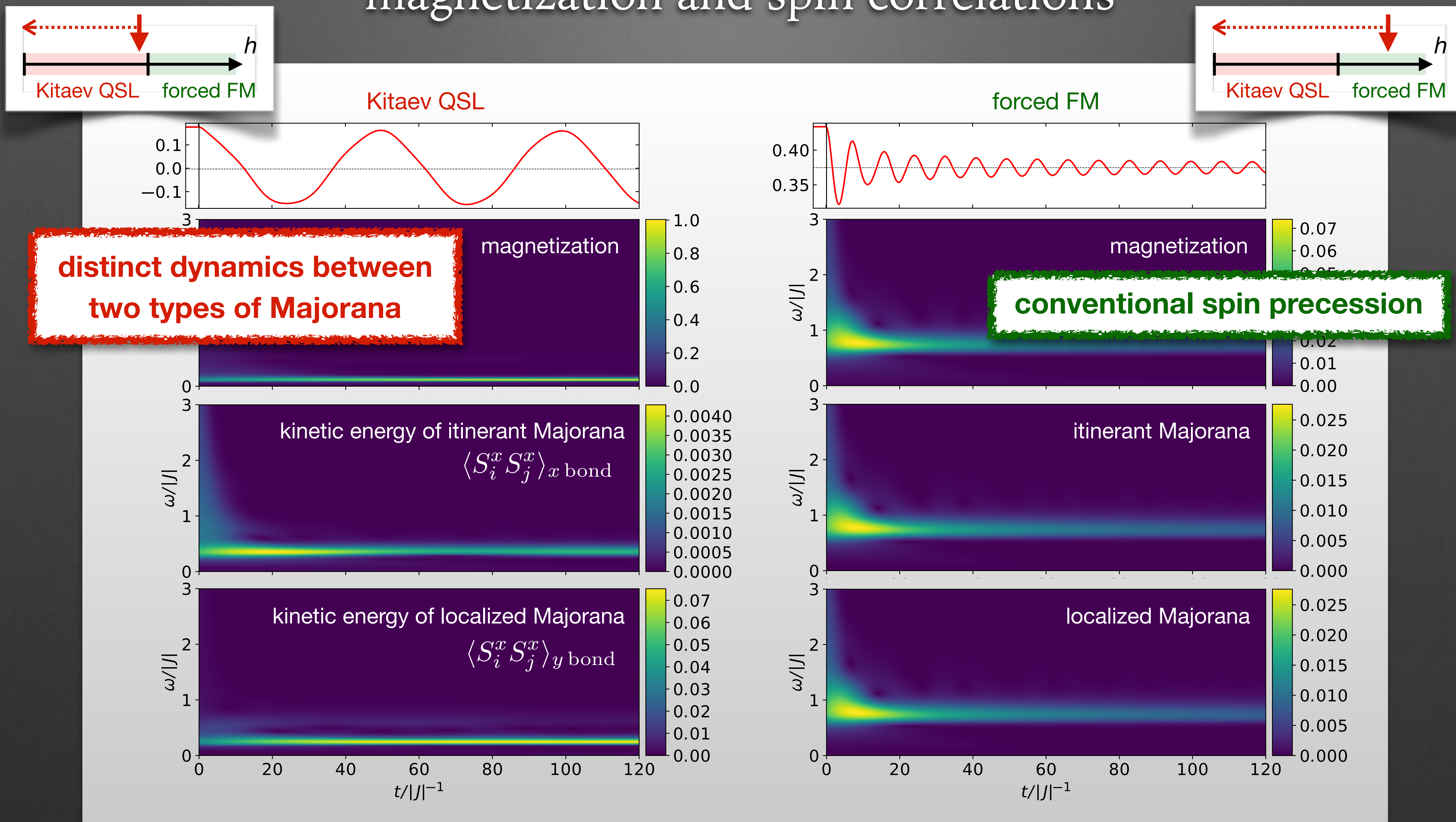
- time-dependent version to study real-time dynamics

$$|\phi_{\mathbf{k}\nu}(t)\rangle = \mathcal{T}\exp\left[-i\int_0^t \mathcal{H}_{\mathbf{k}}^{\mathrm{MF}}(t')dt'\right]|\phi_{\mathbf{k}\nu}(0)\rangle$$

- As a first step, we consider a quench of the magnetic field.

Real-time evolution

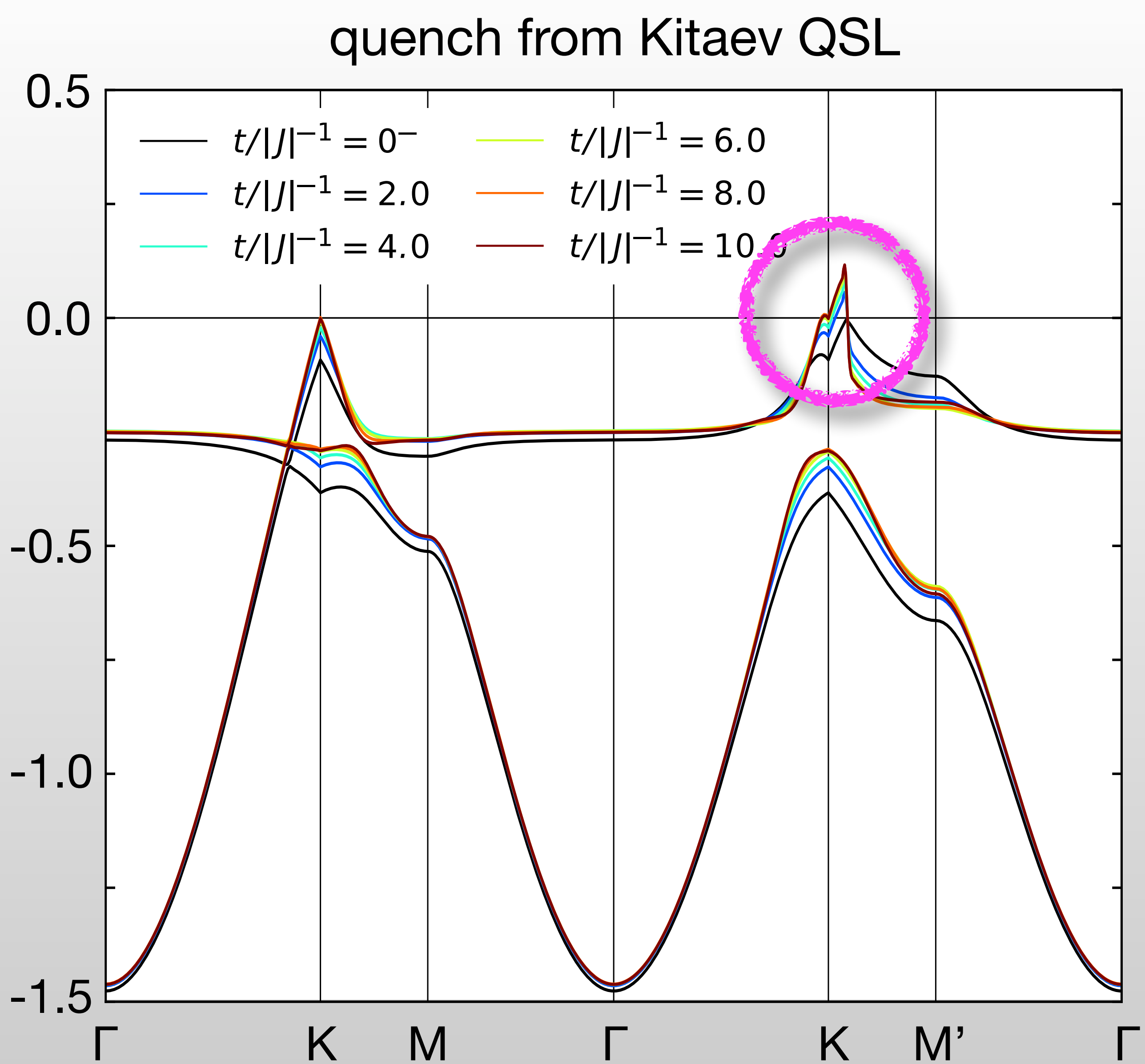
magnetization and spin correlations

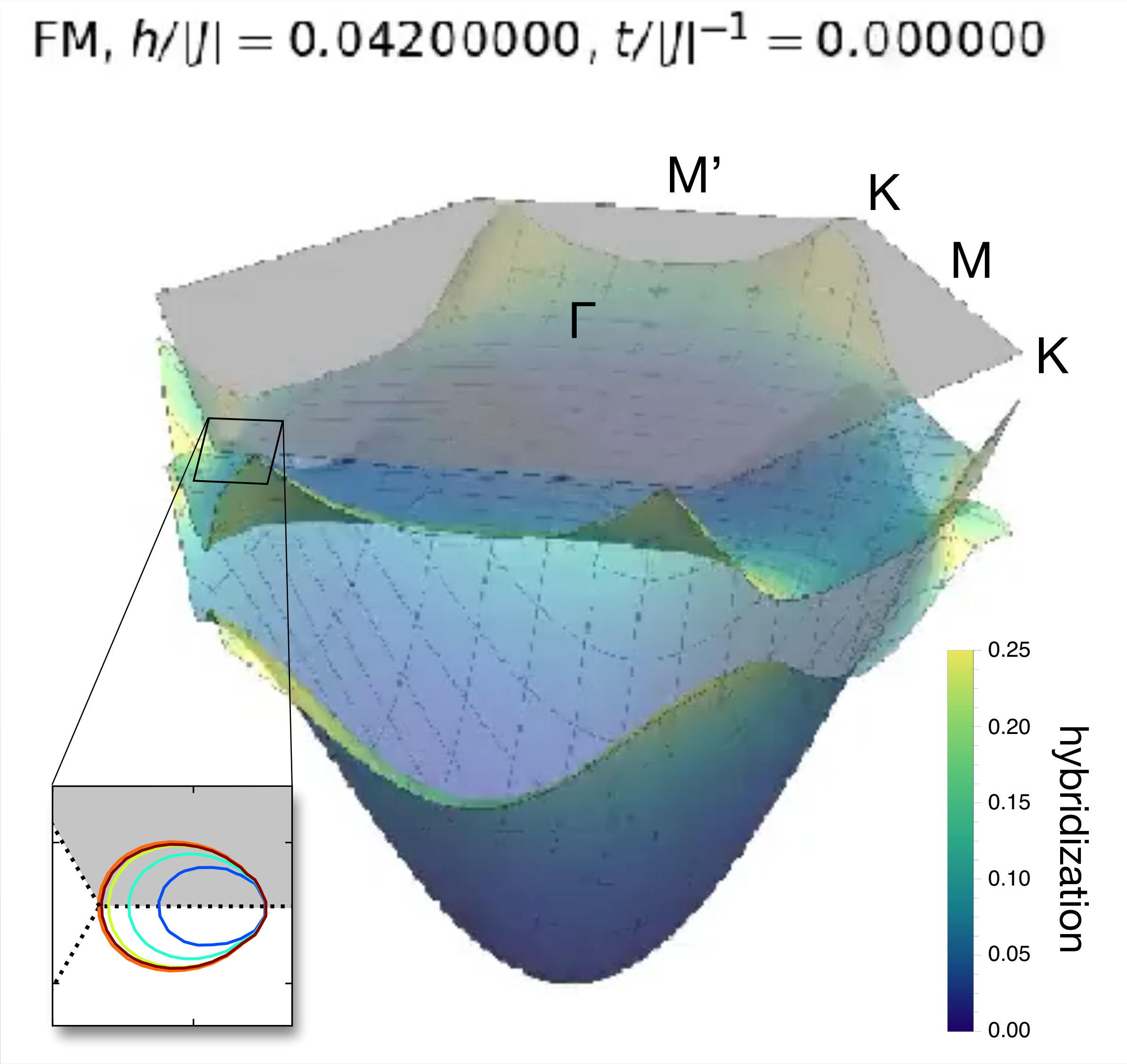


J. Nasu and YM, preprint (arXiv:1905.10984), to appear in Phys. Rev. Research

Majorana band structure

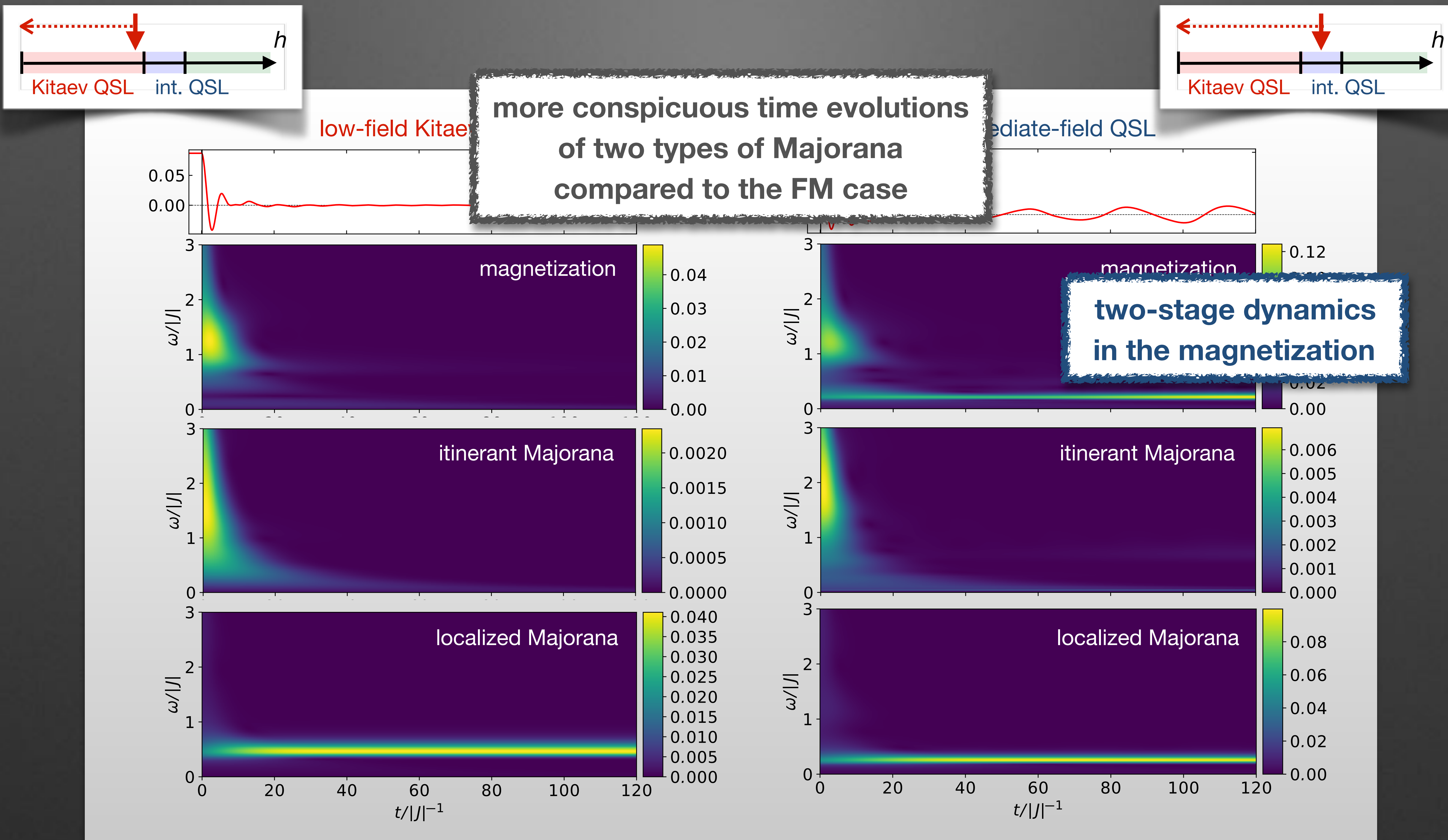
Transient Majorana "Fermi surfaces"





J. Nasu and YM, preprint (arXiv:1905.10984), to appear in Phys. Rev. Research

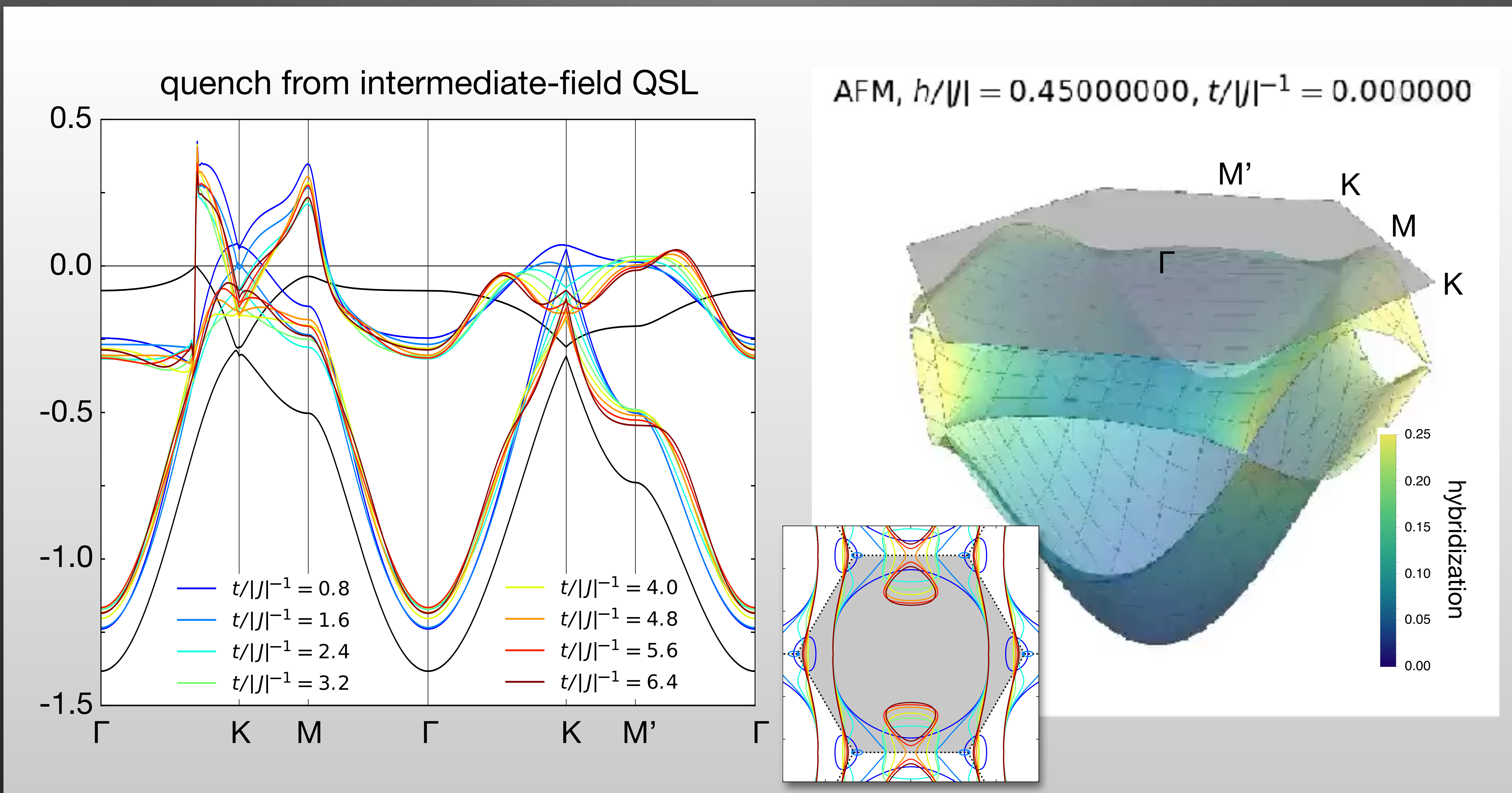
Antiferro Kitaev case



J. Nasu and YM, preprint (arXiv:1905.10984), to appear in Phys. Rev. Research

Transient "Fermi surfaces"

Dynamical "Lifshitz transition"



J. Nasu and YM, preprint (arXiv:1905.10984), to appear in Phys. Rev. Research

Experimental relevance

- **typical timescale of the fractional dynamics ~10-100 ps:** optical techniques, such as Faraday and Kerr effects, might be applicable to the observation
- **useful for identifying fractional excitations and distinguishing topological phases**
- **transient Fermi surfaces:** Peierls instability? hidden phases, such as dimerized phases through the coupling to lattice deformations and symmetry-breaking phases by spontaneous Majorana ordering via quantum many-body effects?

NB. Dissipation is neglected in the present calculations.

J. Nasu and YM, preprint (arXiv:1905.10984), to appear in Phys. Rev. Research

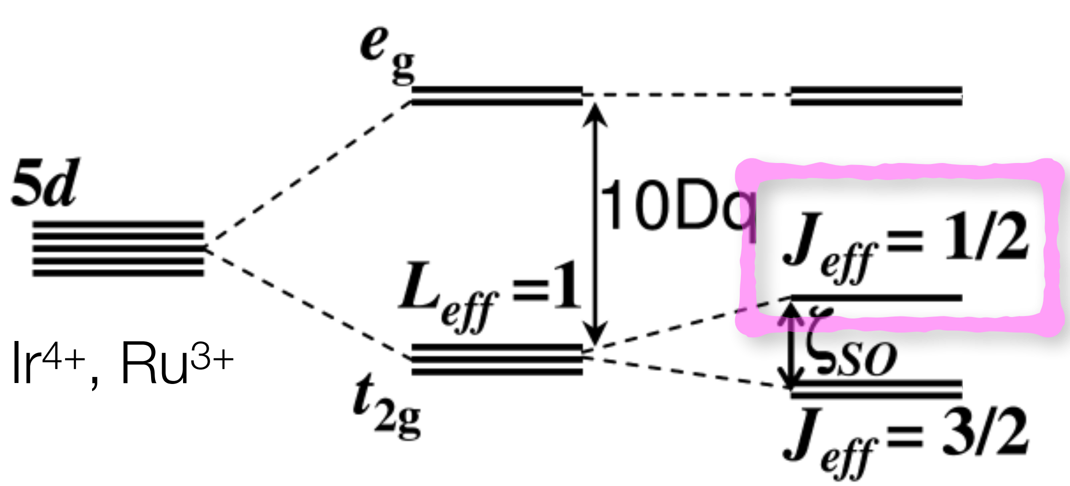
How to materialize Kitaev QSL

Jackeli-Khaliullin mechanism

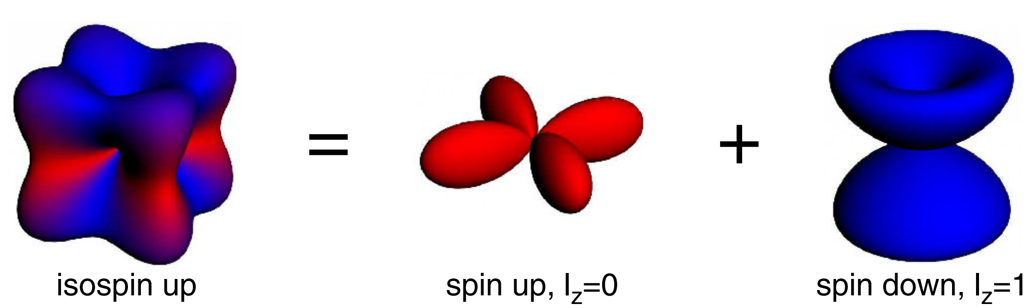
G. Jackeli and G. Khaliullin, Phys. Rev. Lett. **102**, 017205 (2009)

- two requisites for realizing the Kitaev-type anisotropic interactions

spin-orbit entangled Mott insulator with J_{eff}=1/2 Kramers doublet



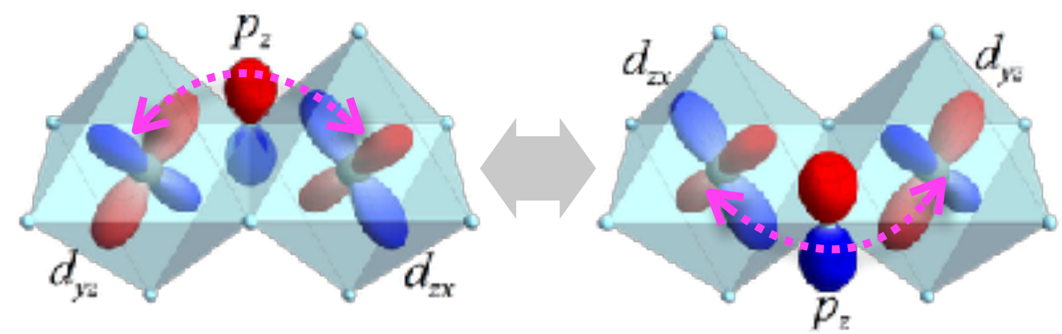
B. J. Kim *et al.*, PRL **101**, 076402 (2008)

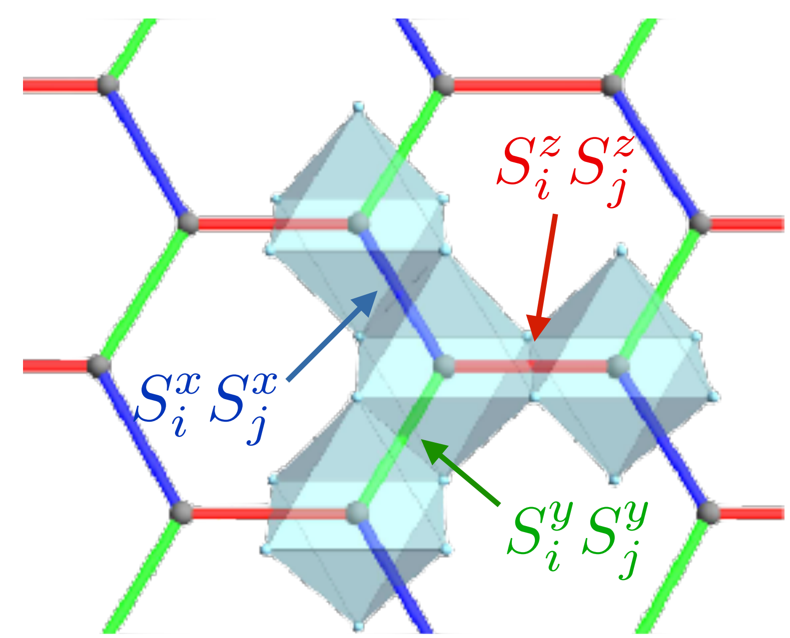


G. Jackeli and G. Khaliullin, PRL **102**, 017205 (2009)

interference between *d*-*p*-*d* transfers

(e.g., edge-sharing octahedra)





Candidate materials

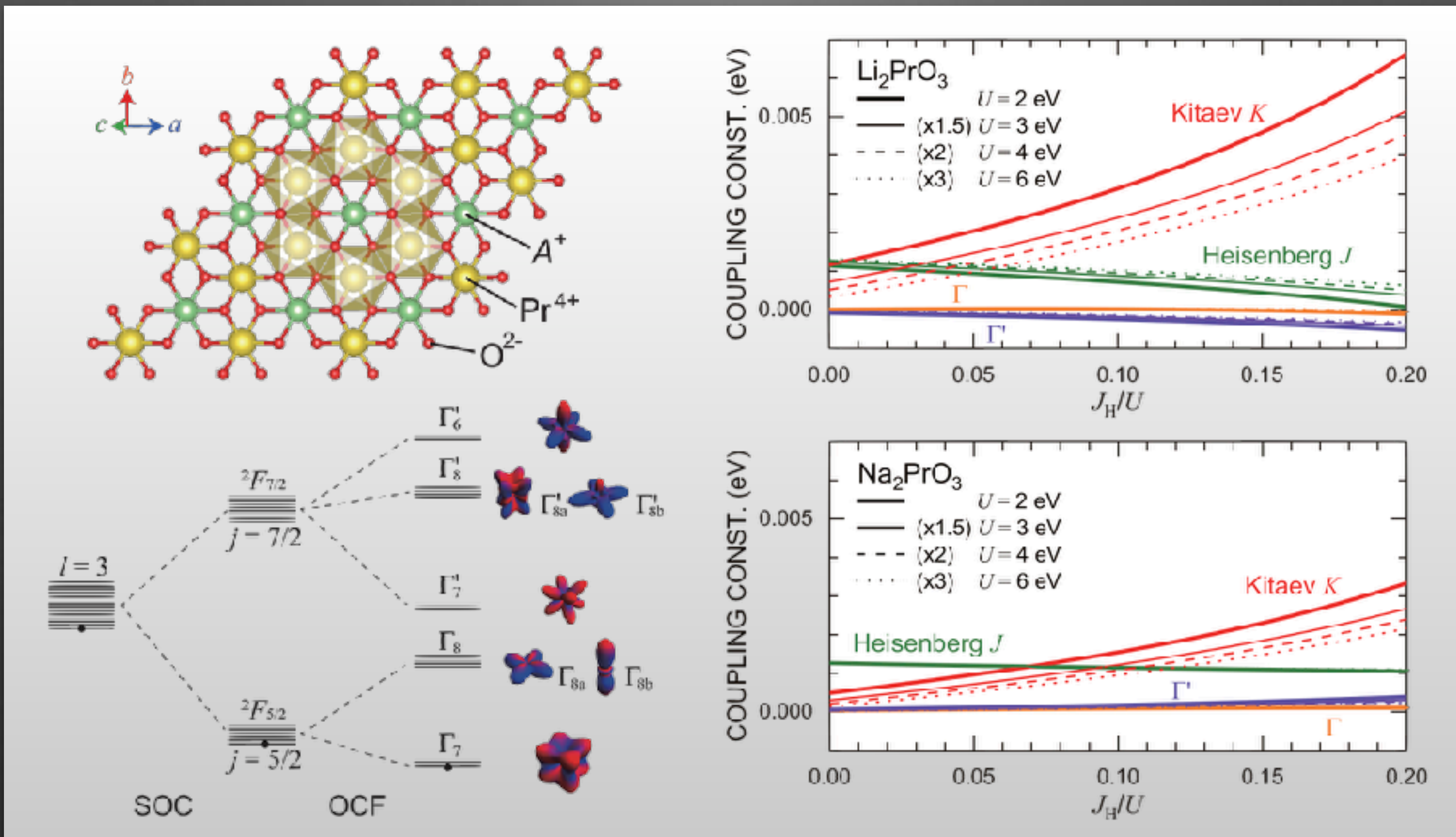
Materials	Crystal structure (space group)	T_{mag}	Anisotropy	p_{eff} (μ_B)	θ_{CW} (K)	Magnetic ground state
Na_2IrO_3	2D ($C2/m$)	15 K	$\chi_c > \chi_{ab}$	1.81 (ab) 1.94 (c)	−176 (θ_{ab}) −40 (θ_c)	Zigzag
α-Li_2IrO_3	2D ($C2/m$)	15 K	$\chi_{ab} > \chi_c$	1.50 (ab) 1.58 (c)	+5 (θ_{ab}), −250 (θ_c)	Spiral
$H_3LiIr_2O_6$	2D ($C2/m$)	–	$\chi_{ab} > \chi_c$	1.60	−105	Spin-liquid
Cu_2IrO_3	[illegible]	[illegible]	[illegible]	[illegible]		Antiferromagnetic order or spin-glass
$Cu_3LiIr_2O_6$	2D ($C2/c$)	15 K	Not known	2.1(1)	−145	Antiferromagnetic order
$Ag_3LiIr_2O_6$	2D (R-3m[a])	~12 K	Not known	1.77		Antiferromagnetic order
α-$RuCl_3$	2D ($C2/m$ or $P3_112$, or R-3); T and sample dependent	7 K and/or 14 K (see text)	$\chi_{ab} > \chi_c$	2.35 (ab), 2.71 (c)	+39.6(θ_{ab}), −216.4 (θ_c)	Zigzag
β-Li_2IrO_3	3D ($Fddd$)	38 K	$\chi_b > \chi_c > \chi_a$	1.87 (a) 1.80 (b) 1.97 (c)	−90.2 (θ_a) +12.9 (θ_b) +21.6 (θ_c)	Spiral
γ-Li_2IrO_3	3D ($Cccm$)	39.5 K	$\chi_b > \chi_c > \chi_a$	~1.6	+40	Spiral

All the existing candidates are believed to have
the ferro Kitaev interactions arising from the J-K mechanism.

Q. how to materialize AFM Kitaev interactions?

H. Takagi, T. Takayama, G. Jackeli, G. Khaliullin, and S. E. Nagler, Nat. Rev. Phys. 1, 264 (2019)

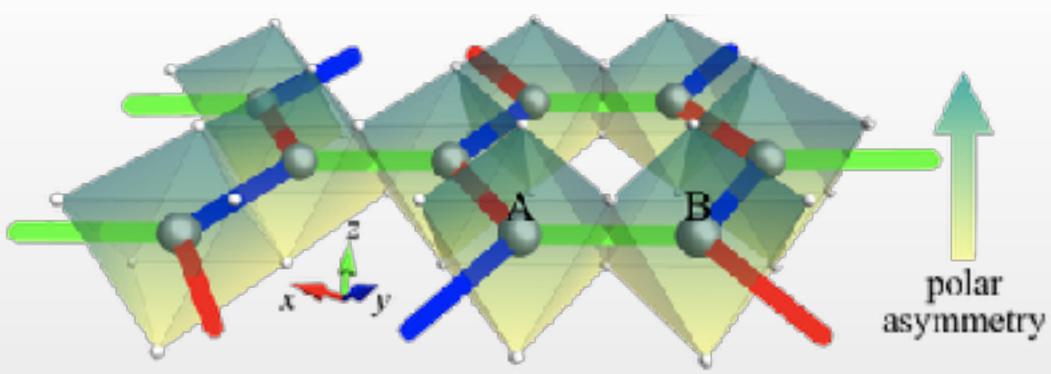
$4f^1$ electron compounds



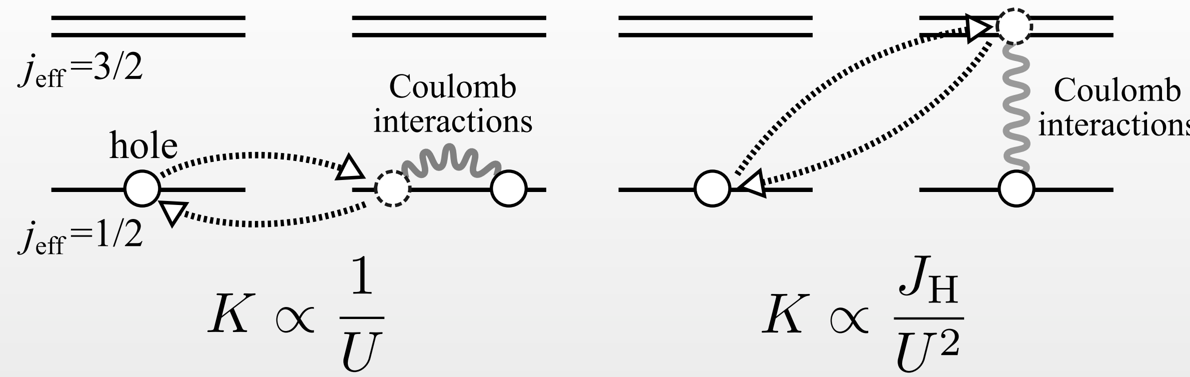
S. Jang, R. Sano, Y. Kato, and YM, Phys. Rev. B 99, 241106(R) (2019)

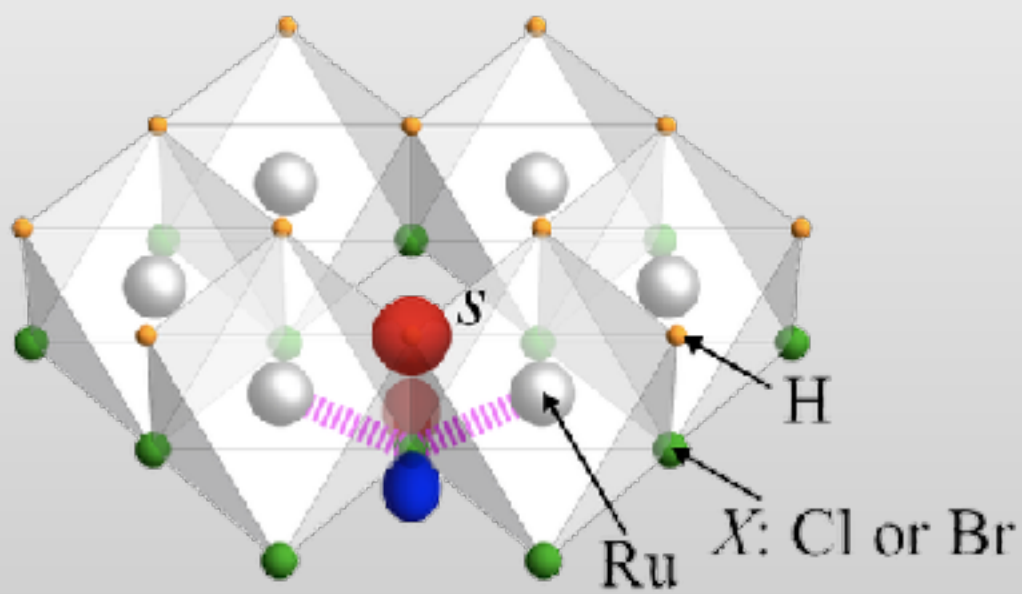
cf. F.-Y. Li, Y.-D. Li, Y. Yu, A. Paramekanti, and G. Chen, Phys. Rev. B 95, 085132; J. G. Rau and M. J. P. Gingras, Phys. Rev. B 98, 054408 (2018)

Polar spin-orbit Mott insulator

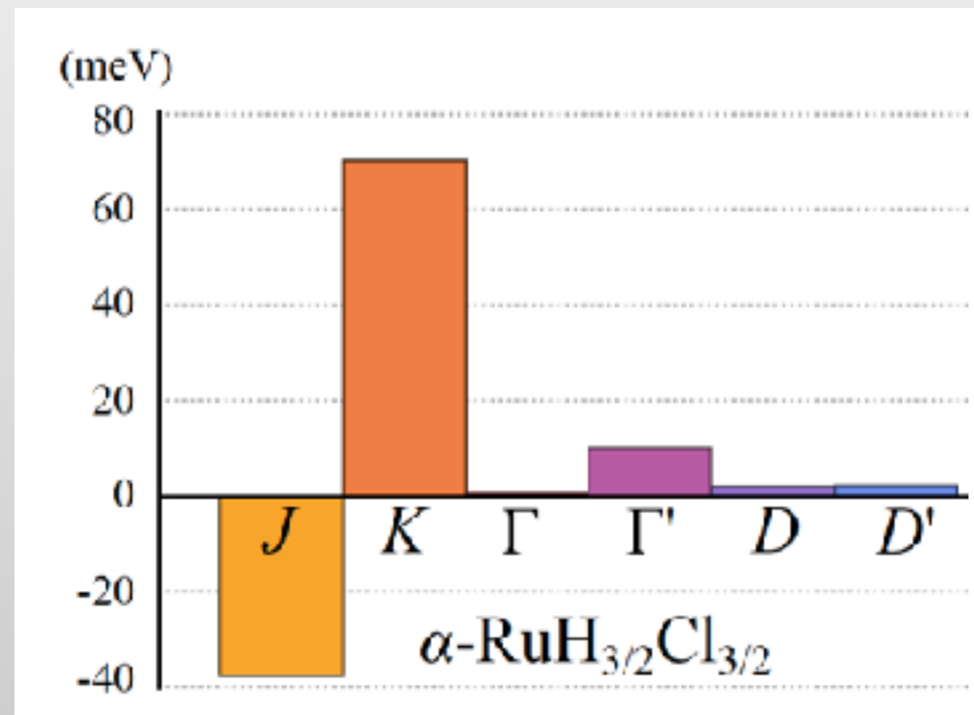


surface/interface, …





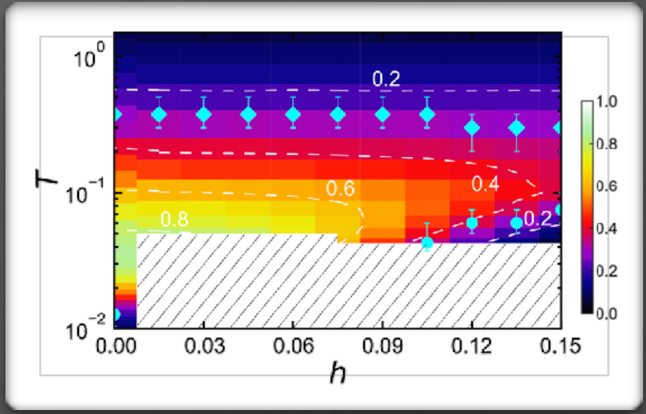
mixed anion



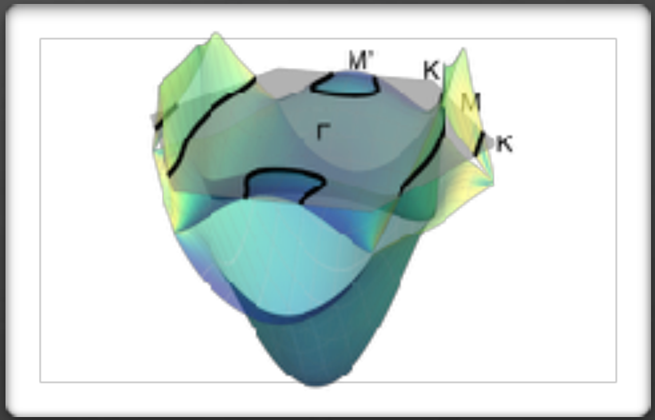
Y. Sugita, Y, Kato, and YM, preprint (arXiv:1905.12139)

Summary and perspectives

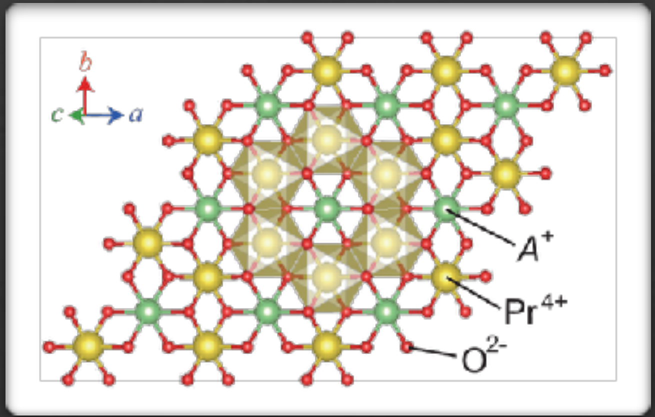
- **Signatures of fractional excitations in a magnetic field at finite temperature *T***
  - extremely wide fractional state beyond the critical field at T=0
  - Majorana-magnon crossover: confinement-deconfinement
  - ➡ further comparison between theory and experiments, more sophisticated theory for lower T in the field, …



- **Real-time dynamics of fractional excitations by magnetic field quench**
  - distinct time evolution of two types of fractional excitations
  - transient Majorana fermi surfaces: dynamical Lifshitz transition
  - ➡ experimental confirmation, other real-time dynamics, …



- **How to materialize the Kitaev model with antiferromagnetic Kitaev interactions**
  - $4f^1$ electron compounds A_2PrO_3
  - polar spin-orbit Mott insulators
  - ➡ experimental confirmation, other sorts of candidates, …



Collaborators

My group (Tokyo):

Yasuyuki Kato   Troels Bojesen   Petr Mishchenko   Junki Yoshitake
Ryoya Sano   Seonghoon Jang   Yusuke Sugita

Former members of my group:

Joji Nasu (Yokohama)   **Masafumi Udagawa** (Gakushuin)

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