

Majorana-fermion origin for the thermal Hall effect of Kitaev candidate α -RuCl₃



Yuji Matsuda

Department of Physics, Kyoto University, Japan



大



Majorana-fermion origin for the thermal Hall effect of Kitaev candidate α -RuCl₃

Y. Kasahara, S. Suetsugu, T. Asaba, Y. Xing,
T. Yokoi, S. Kasahara, Y. Kohsaka
Department of Physics, Kyoto Univ., Japan



O. Tanaka, K. Imamura, K. Hashimoto, T. Shibauchi
Department of Adv. Materials Science, Univ. of Tokyo, Japan



N. Kurita, H. Tanaka

Department of Physics, Tokyo Institute of Technology, Japan

Y. Mizukami, J. Nasu

Department of Physics, Tohoku University, Japan



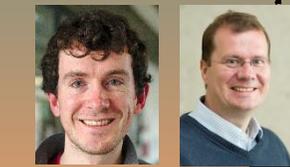
Y. Motome

Department of Applied Physics, Univ. of Tokyo, Japan



M. G. Yamada, S. Fujimoto

Department of Materials Engineering Science, Osaka Univ., Japan

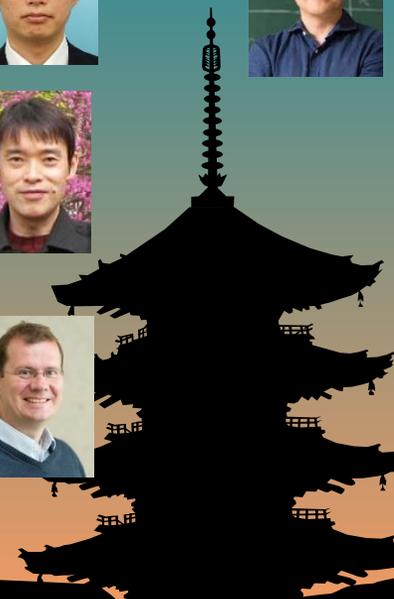


E-G. Moon

KAIST, Korea

C. Hickey, S. Trebst

Department of Physics, Univ. of Cologne, Germany



Contents

1. Half integer quantized thermal Hall effect in α - RuCl_3

2. Is α - RuCl_3 really Kitaev material?

Topology: Topological invariant

Thermodynamics: Dirac cone and Majorana gap

Thermal transport properties of very clean crystals

3. STM studies on monolayer α - RuCl_3

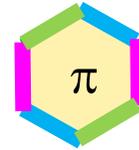
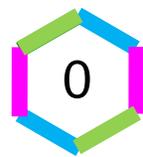
Local density of states around defects

Kitaev quantum spin liquid

Spin 1/2

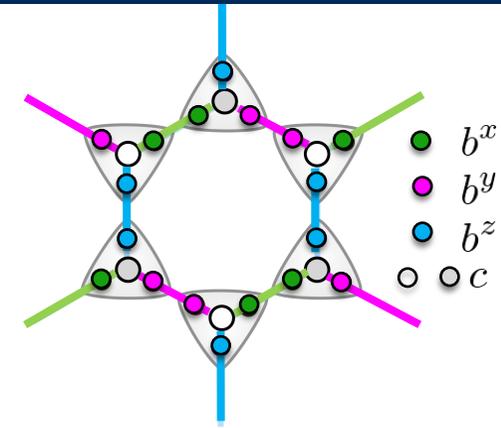
Itinerant Majorana c

Localized Majorana b^x, b^y, b^z



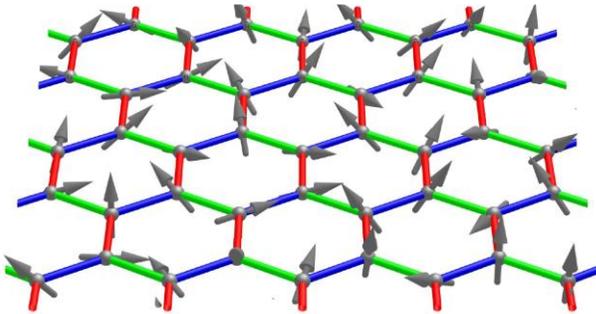
Z_2 gauge flux (Vison)

Energy gap: $\Delta_F \sim 0.1J_K$



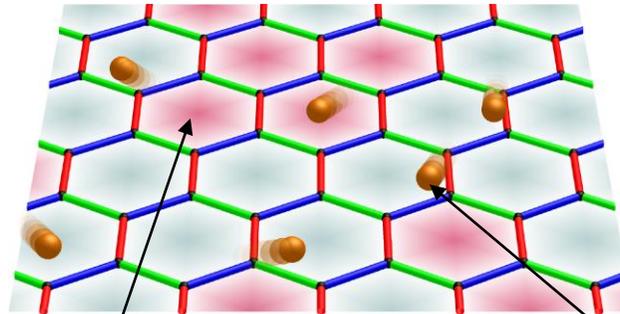
$T > J_K$

Paramagnetic



$T < J_K$

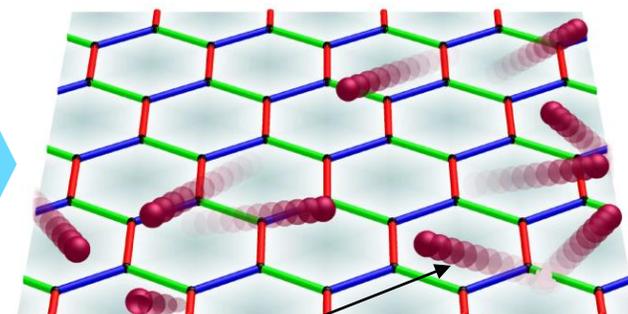
Spin fractionalization



Z_2 flux

$T \ll J_K$

Quantum spin liquid



Itinerant Majorana fermions

J. Nasu and Y. Motome

Strongly spin-orbit coupled $j=1/2$ Mott insulator α -RuCl₃

A prime candidate for hosting an approximate Kitaev QSL

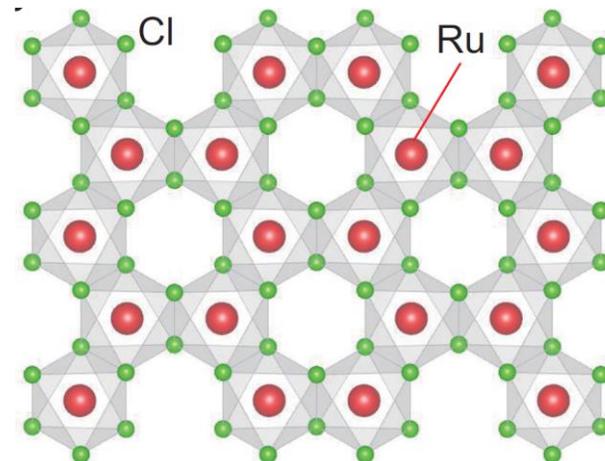
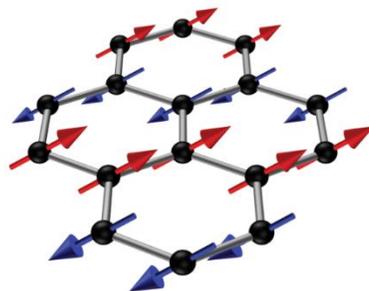
$$\mathcal{H}_{ij}^{(z)} = \underbrace{K S_i^z S_j^z}_{\text{Kitaev}} + \underbrace{J \mathbf{S}_i \cdot \mathbf{S}_j}_{\text{Heisenberg}} + \underbrace{\Gamma (S_i^x S_j^y + S_i^y S_j^x)}_{\text{off-diagonal}} + \underbrace{\Gamma' (S_i^x S_j^z + S_i^z S_j^x + S_i^y S_j^z + S_i^z S_j^y)}_{\text{off-diagonal (distortion)}}$$

Significant Kitaev term

RIXS $K = -5 \text{ meV}$ Ferromagnetic
 $J = -3 \text{ meV}$ $\Gamma = 2.5 \text{ meV}$ $\Gamma' \approx 0.1 \text{ meV}$
 H. Suzuki et al. Nature Commun. **12**, 4512 (2021).

Non-Kitaev interaction

AFM order at $T_N = 7.5 \text{ K}$



K.W. Plumb *et al.* PRB **90**, 041112 (2014).

Strongly spin-orbit coupled $j=1/2$ Mott insulator α -RuCl₃

A prime candidate for hosting an approximate Kitaev QSL

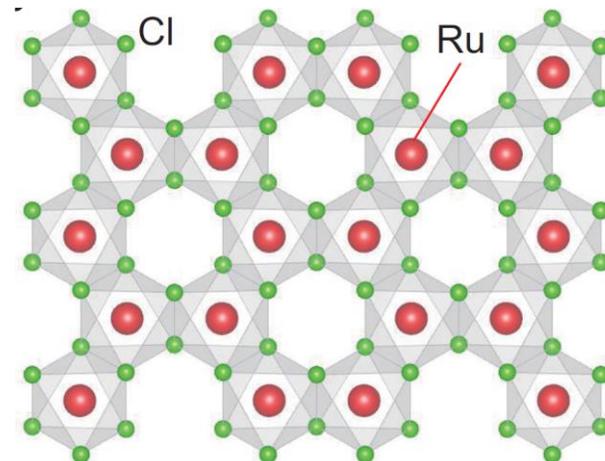
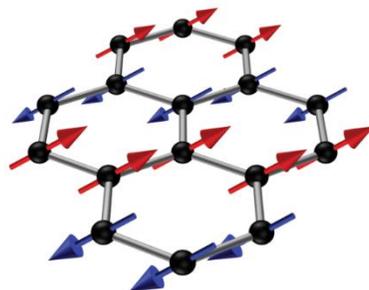
$$\mathcal{H}_{ij}^{(z)} = \underbrace{K S_i^z S_j^z}_{\text{Kitaev}} + \underbrace{J \mathbf{S}_i \cdot \mathbf{S}_j}_{\text{Heisenberg}} + \underbrace{\Gamma (S_i^x S_j^y + S_i^y S_j^x)}_{\text{off-diagonal}} + \underbrace{\Gamma' (S_i^x S_j^z + S_i^z S_j^x + S_i^y S_j^z + S_i^z S_j^y)}_{\text{off-diagonal (distortion)}}$$

Significant Kitaev term

RIXS $K = -5 \text{ meV}$ Ferromagnetic
 $J = -3 \text{ meV}$ $\Gamma = 2.5 \text{ meV}$ $\Gamma' \approx 0.1 \text{ meV}$
 H. Suzuki *et al.* Nature Commun. **12**, 4512 (2021).

Non-Kitaev interaction

AFM order at $T_N = 7.5 \text{ K}$



K.W. Plumb *et al.* PRB **90**, 041112 (2014).

Signatures of fractionalization of electron spin

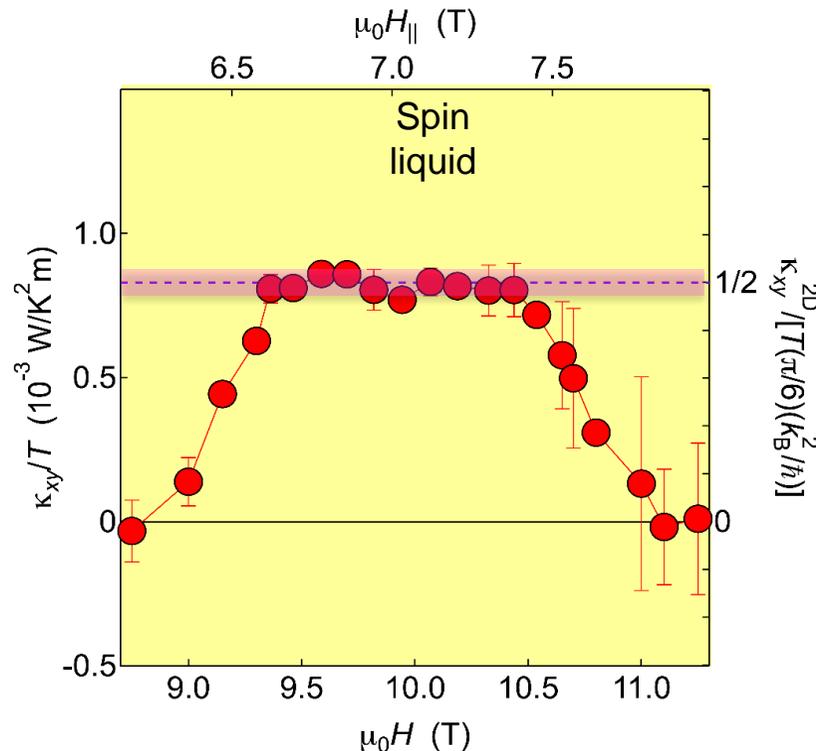
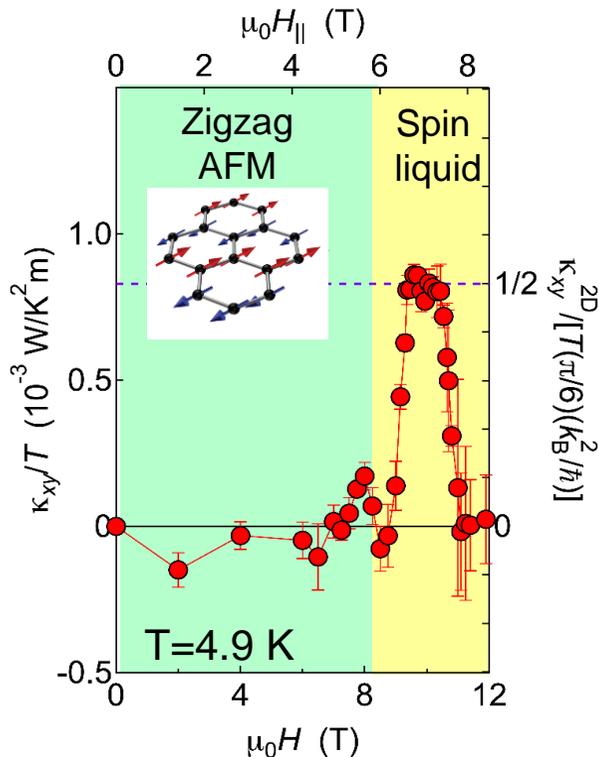
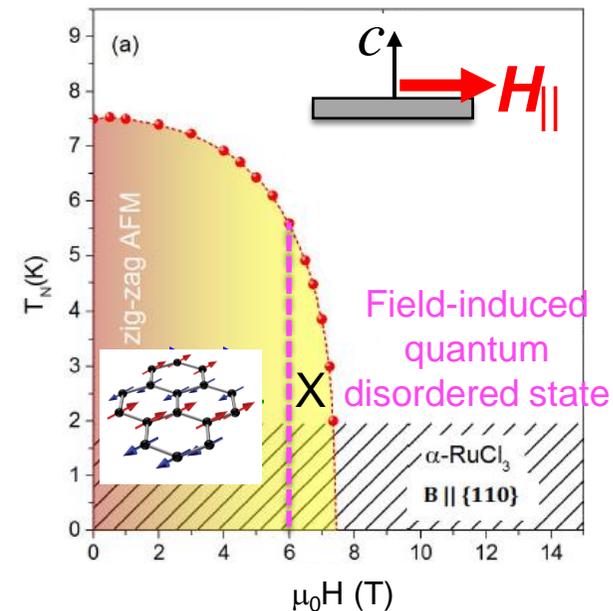
Specific heat	Entropy release due to itinerant Majorana fermions S-H.Do <i>et al.</i> Nature Phys. 13 , 1079 (2017). S. Widmann <i>et al.</i> PRB 99 , 094415 (2019).
Raman	Fermionic excitations inconsistent with bosonic magnons J. Nasu <i>et al.</i> , Nature Phys. 12 , 912 (2016).
Neutron	Broad magnetic continuum in good agreement with Kitaev QSL A. Banerjee <i>et al.</i> , Nature Mater. 15 , 733 (2016). A. Banerjee <i>et al.</i> , Science 356 , 1055 (2017).

Half-integer quantized thermal Hall conductance plateau

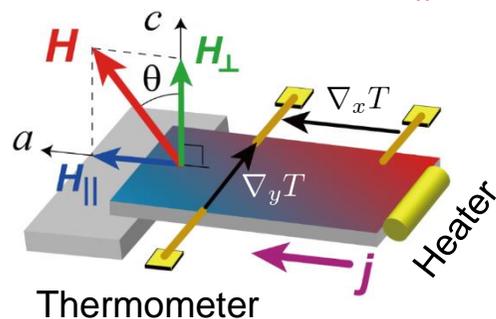
A. Banerjee *et al.*, npj Quantum Mat. **3**, 8 (18).

Y. Kasahara, YM, *et al.*
Nature **559**, 227 (2018).

$$\theta = -45^\circ$$



Zigzag AFM order is suppressed by H_{\parallel}



Thermal Hall effect in tilted fields

$$\kappa_{xy}^{2D} = \frac{1}{2} K_0$$

$$K_0 = \frac{\pi^2 k_B^2}{3h} T$$

Quantum Thermal Conductance

Thermal Hall plateau

Majorana edge current

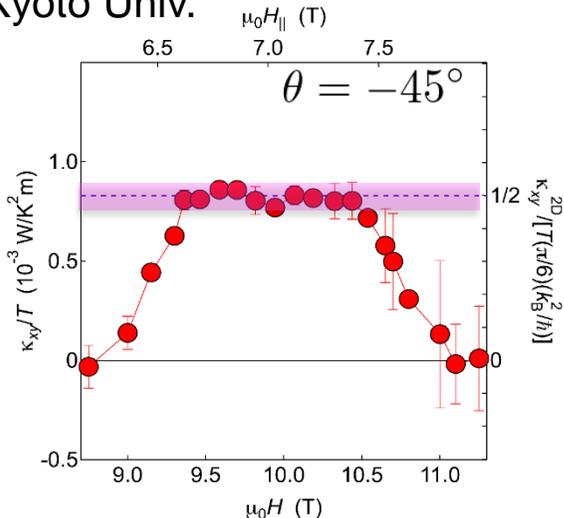
Non-abelian anyon

Reproducibility of thermal Hall effect

1. Quantized plateau

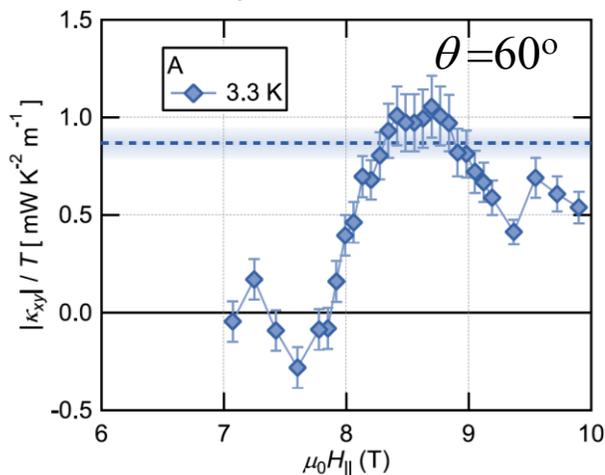
Crystals grown by Bridgman method

Kyoto Univ.



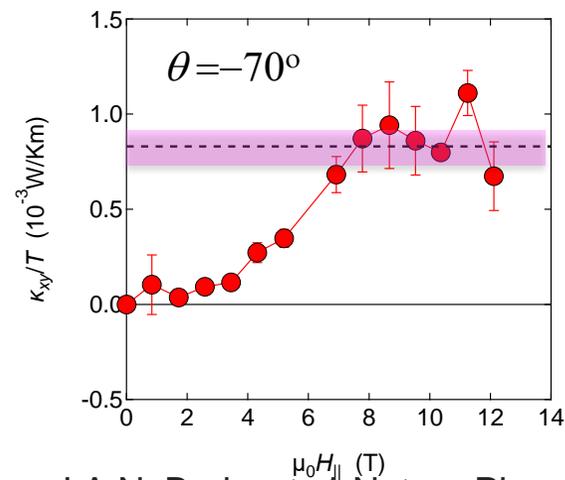
Y. Kasahara *et al.* Nature (2018).

Univ. of Tokyo



M. Yamashita *et al.* PRB (2020).

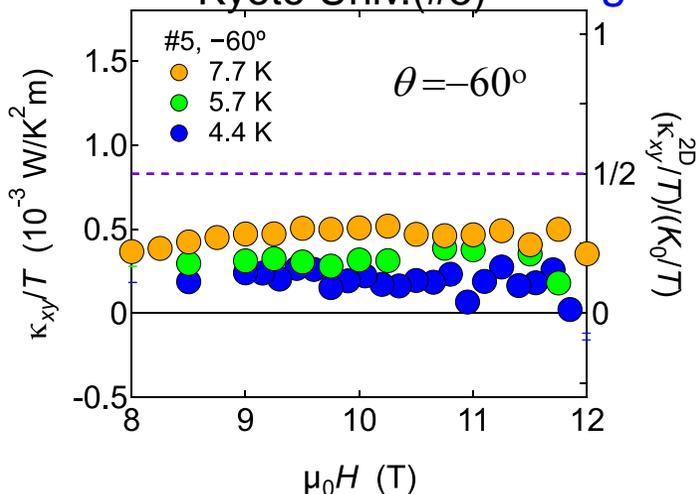
MPI Stuttgart



J.A.N. Bruin *et al.* Nature Phys. (2022).

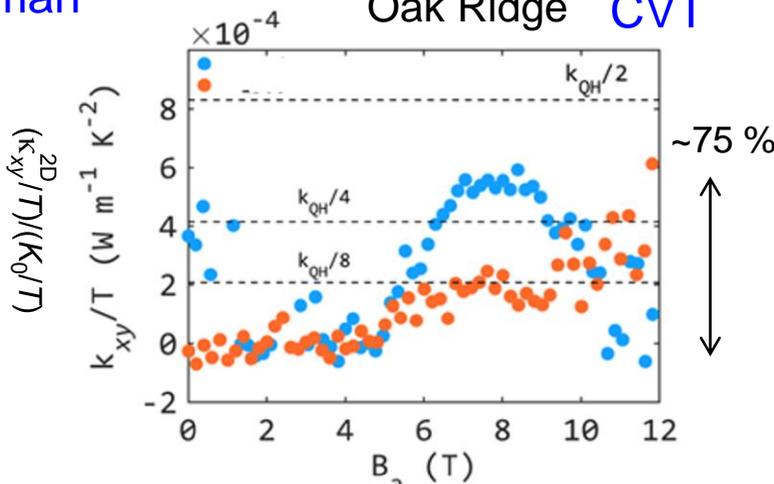
2. Plateau but no quantization

Kyoto Univ. (#5) Bridgman



Y. Kasahara *et al.* PRB (2022).

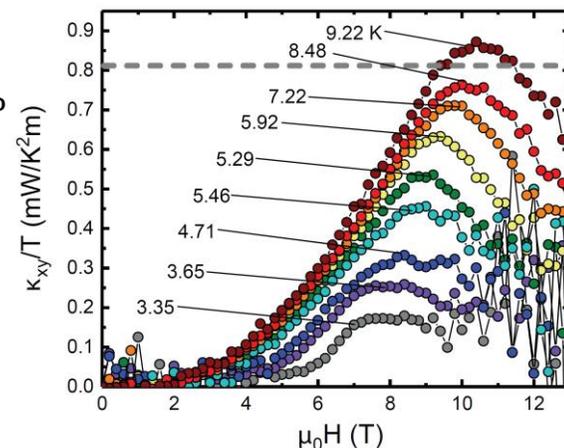
Oak Ridge CVT



H. Zhang *et al.* arXiv:2303.03682

3. No plateau and no quantization

Princeton Univ. CVT



P. Czajka *et al.*, Nature Mater. (2022)

1. Half integer quantized thermal Hall effect in α - RuCl_3

2. Is α - RuCl_3 really Kitaev material?

Topology: Topological invariant

Thermodynamics: Dirac cone and Majorana gap

Thermal transport properties of very clean crystals

3. STM studies on monolayer α - RuCl_3

Local density of states around defects

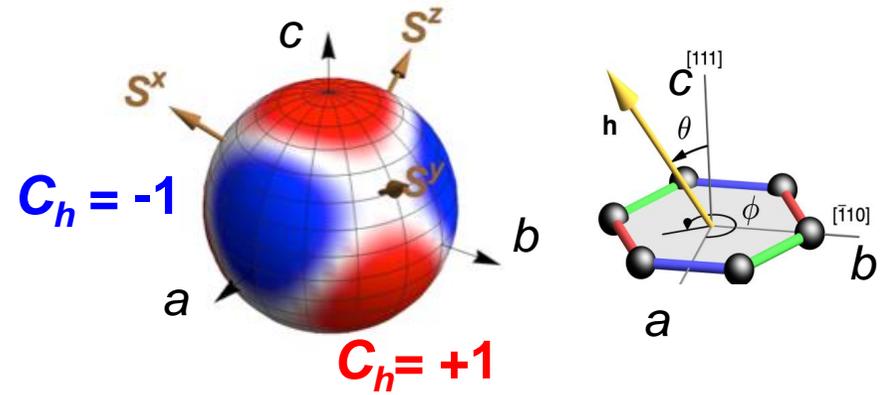
Topological invariant and planar thermal Hall effect

Topological Chern number

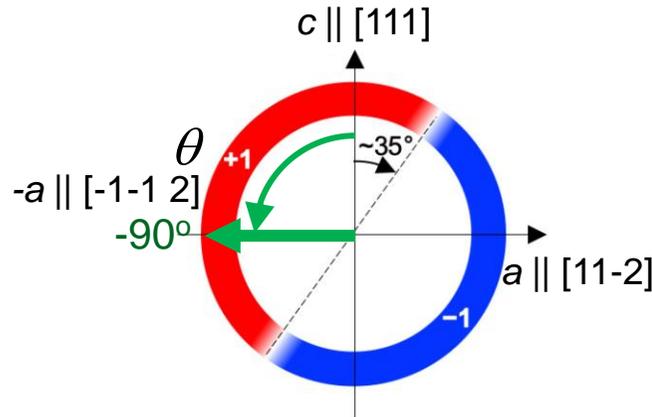
$$\kappa_{xy}^{2D} = C_h \frac{1}{2} K_0 \quad C_h = \text{sgn}(h_x h_y h_z) = \pm 1$$

H-component with respect to the spin axis

Spin axis is different from crystal axis



ac-plane



Finite Chern number in parallel field ($H \parallel -a$, $C_h = +1$)
 Emergence of **planar thermal Hall effect** in paramagnetic state

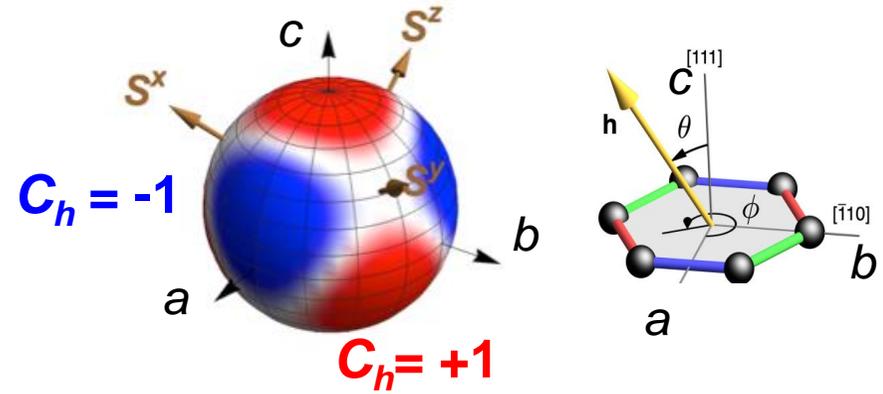
Topological invariant and planar thermal Hall effect

Topological Chern number

$$\kappa_{xy}^{2D} = C_h \frac{1}{2} K_0 \quad C_h = \text{sgn}(h_x h_y h_z) = \pm 1$$

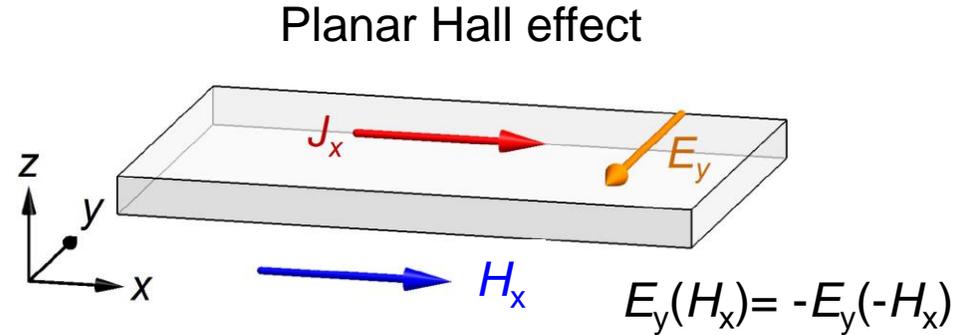
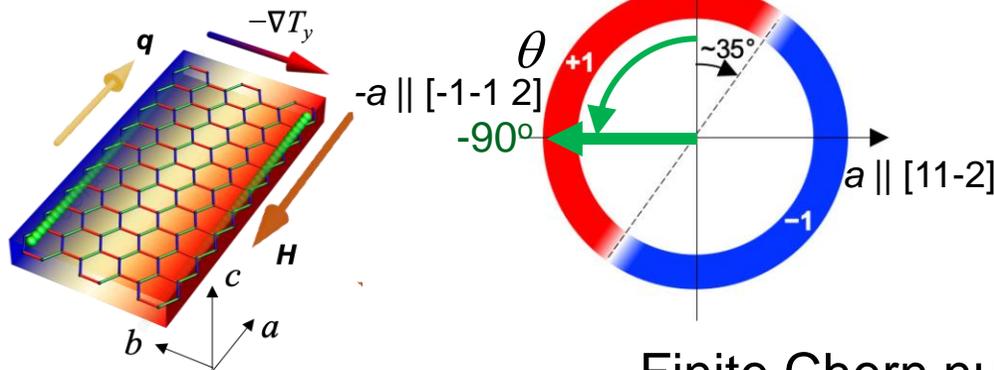
H-component with respect to the spin axis

Spin axis is different from crystal axis



ac-plane

Planar thermal Hall effect



Finite Chern number in parallel field ($H \parallel -a$, $C_h = +1$)

Emergence of **planar thermal Hall effect** in paramagnetic state

$$\nabla T_y(H_x) = -\nabla T_y(-H_x)$$

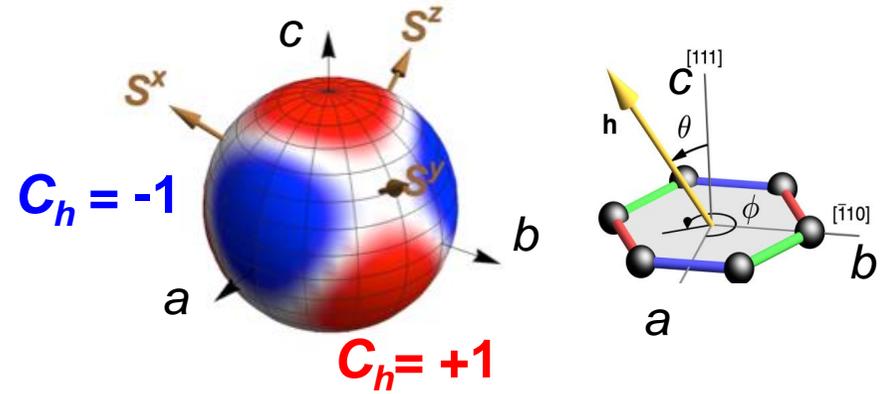
Topological invariant and planar thermal Hall effect

Topological Chern number

$$\kappa_{xy}^{2D} = C_h \frac{1}{2} K_0 \quad C_h = \text{sgn}(h_x h_y h_z) = \pm 1$$

H-component with respect to the spin axis

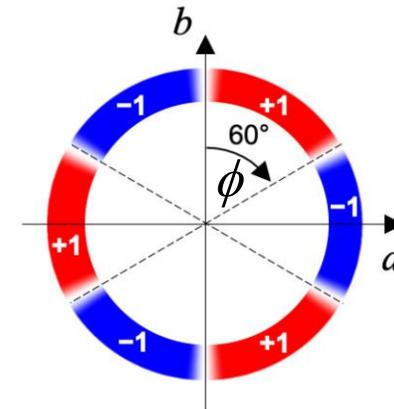
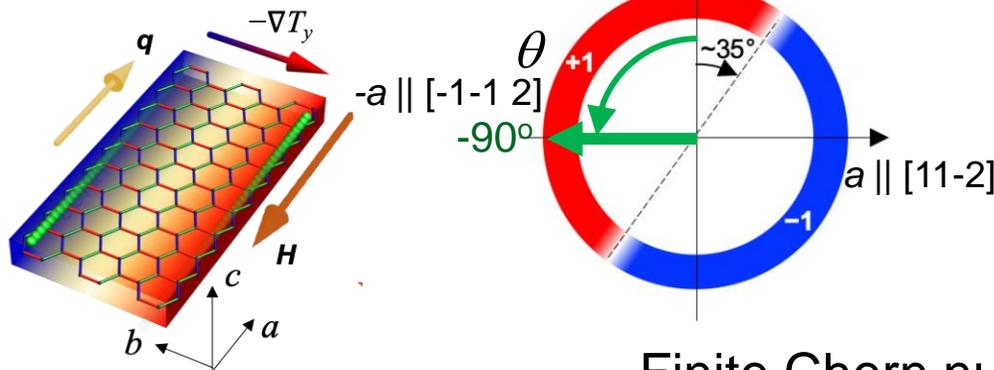
Spin axis is different from crystal axis



ac-plane

ab-plane

Planar thermal Hall effect



Finite Chern number in parallel field ($H \parallel -a$, $C_h = +1$)

Emergence of **planar thermal Hall effect** in paramagnetic state

$$\nabla T_y(H_x) = -\nabla T_y(-H_x)$$

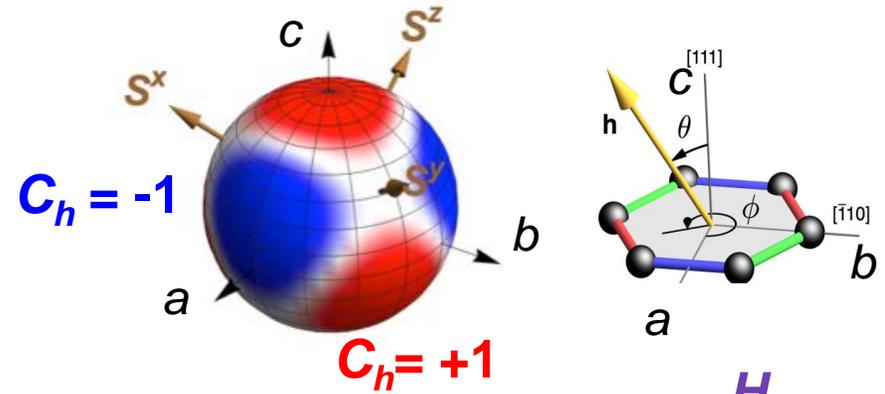
Topological invariant and planar thermal Hall effect

Topological Chern number

$$\kappa_{xy}^{2D} = C_h \frac{1}{2} K_0 \quad C_h = \text{sgn}(h_x h_y h_z) = \pm 1$$

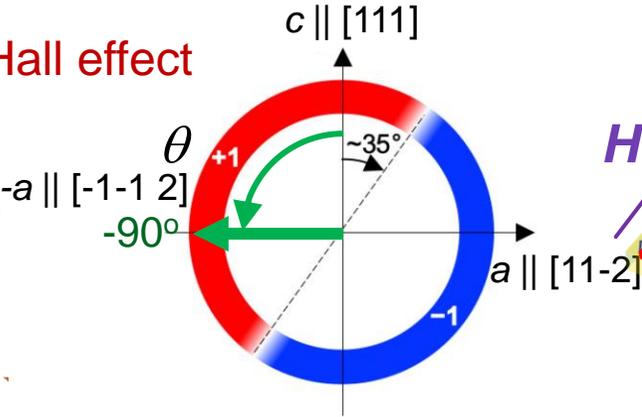
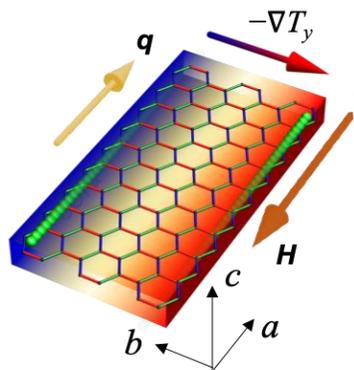
H-component with respect to the spin axis

Spin axis is different from crystal axis

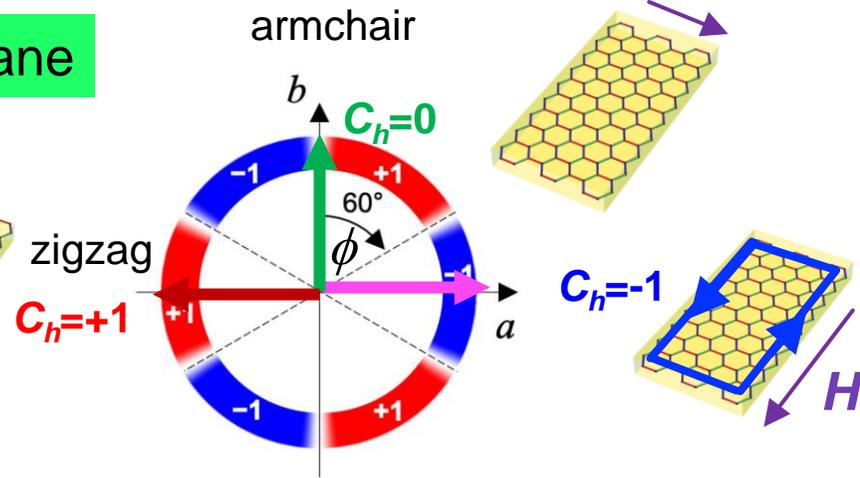
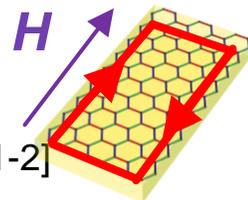


ac-plane

Planar thermal Hall effect



ab-plane



Finite Chern number in parallel field ($H \parallel -a$, $C_h = +1$)

Emergence of **planar thermal Hall effect** in paramagnetic state

$$\nabla T_y(H_x) = -\nabla T_y(-H_x)$$

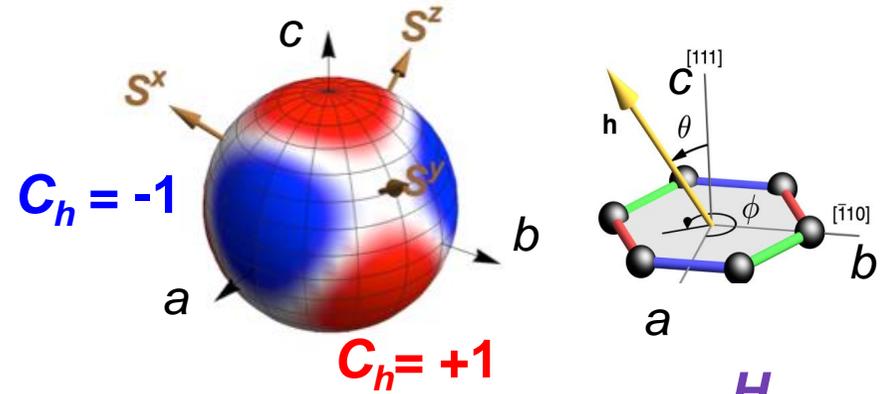
Topological invariant and planar thermal Hall effect

Topological Chern number

$$\kappa_{xy}^{2D} = C_h \frac{1}{2} K_0 \quad C_h = \text{sgn}(h_x h_y h_z) = \pm 1$$

H-component with respect to the spin axis

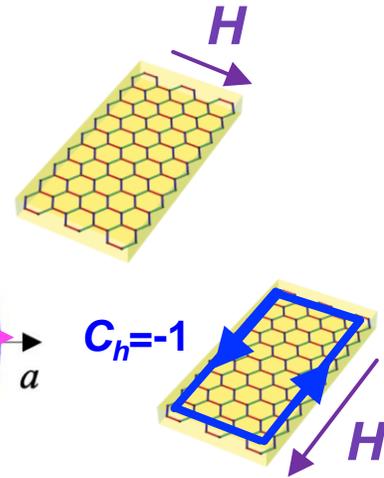
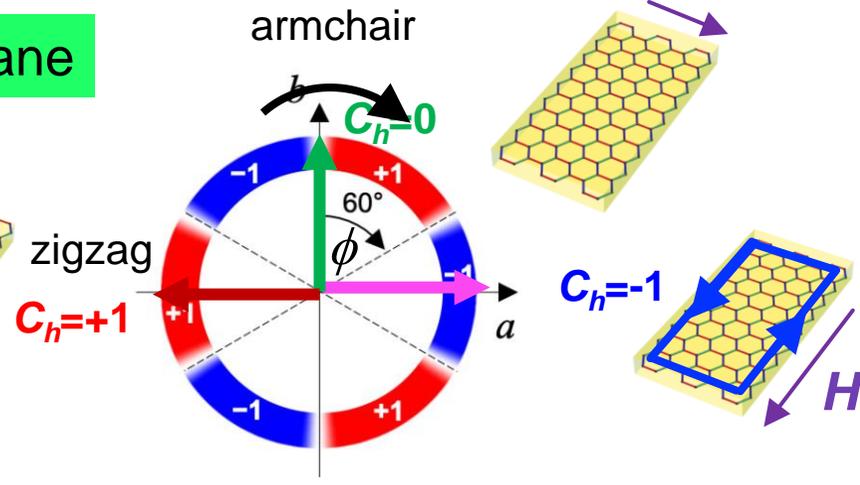
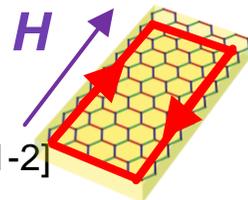
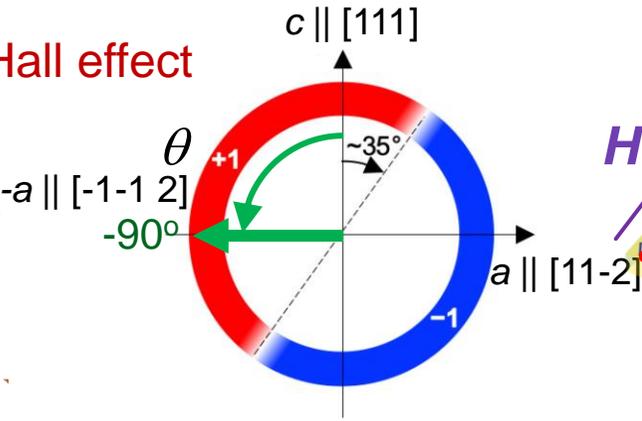
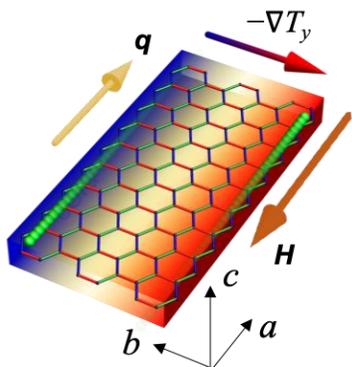
Spin axis is different from crystal axis



ac-plane

ab-plane

Planar thermal Hall effect



$$\nabla T_y(H_x) = -\nabla T_y(-H_x)$$

Finite Chern number in parallel field ($H \parallel -a$, $C_h = +1$)

Emergence of **planar thermal Hall effect** in paramagnetic state

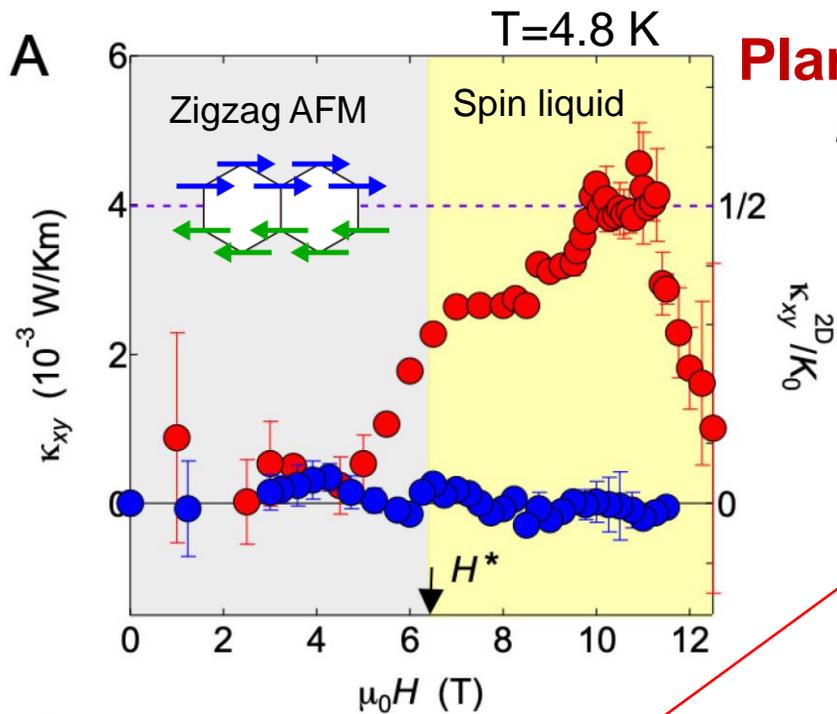
Angle-rotation induced topological transition

Rotating **H** clockwise changes the thermal Hall sign from negative to positive when **H** crosses the **b** axis.

Half-integer thermal QHE in in-plane magnetic field

T. Yokoi, YM *et al.* Science **373**, 568 (2021).

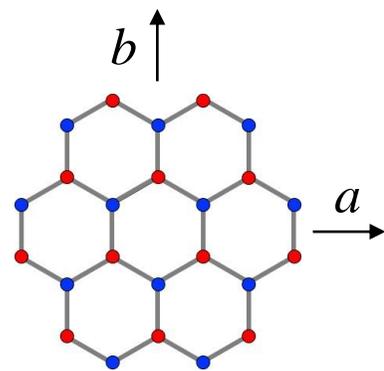
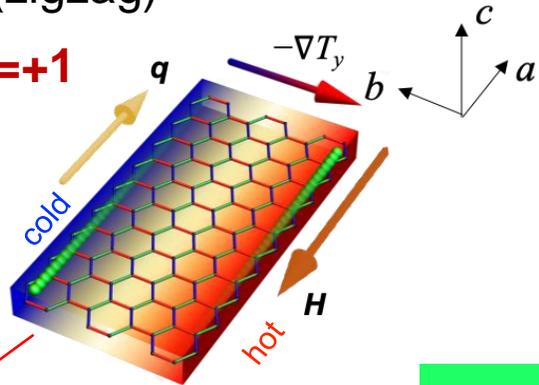
A



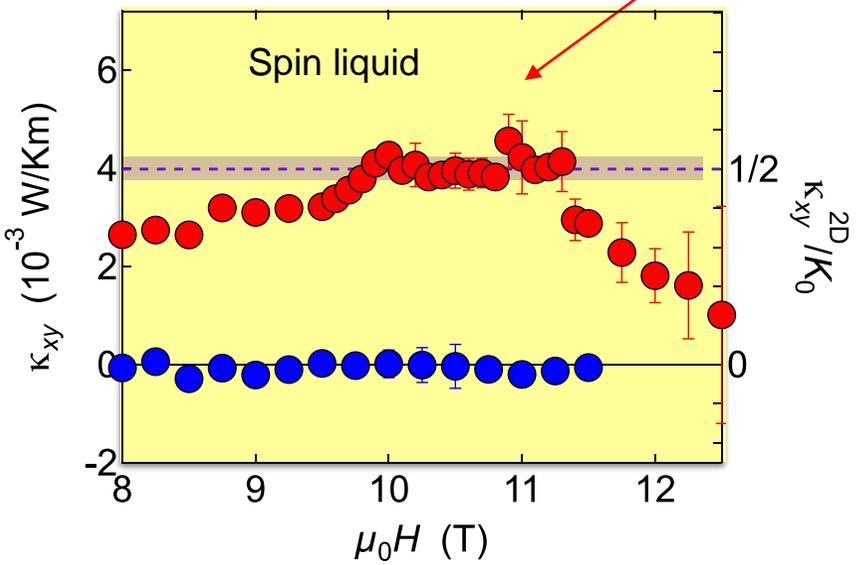
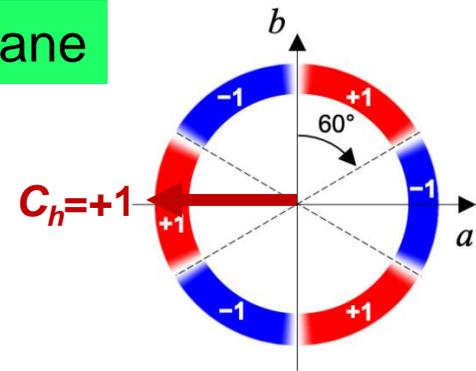
Planar thermal Hall effect

$H // -a$ (zigzag)

$C_h = +1$

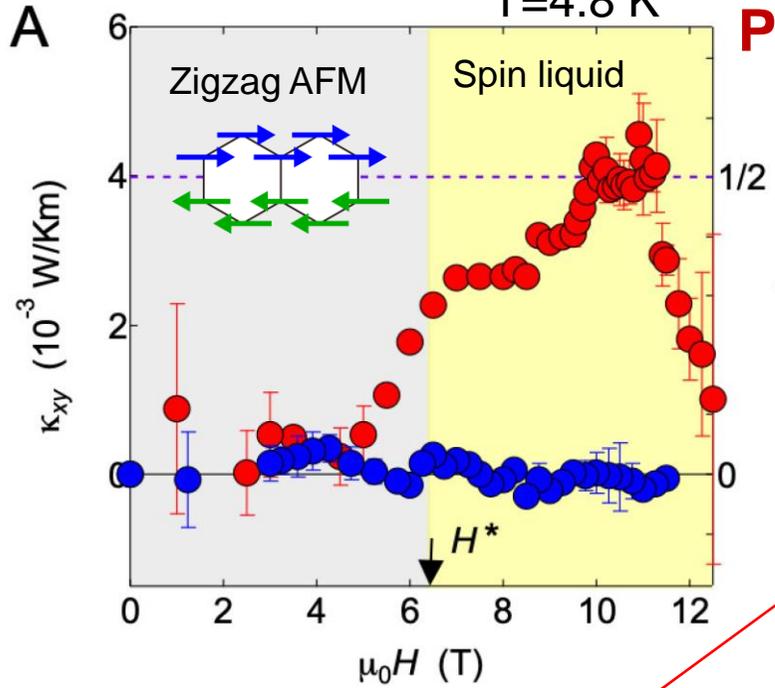


ab-plane



Half-integer thermal QHE in in-plane magnetic field

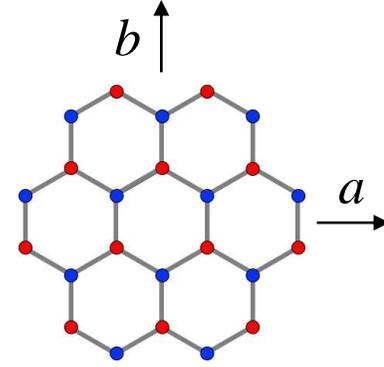
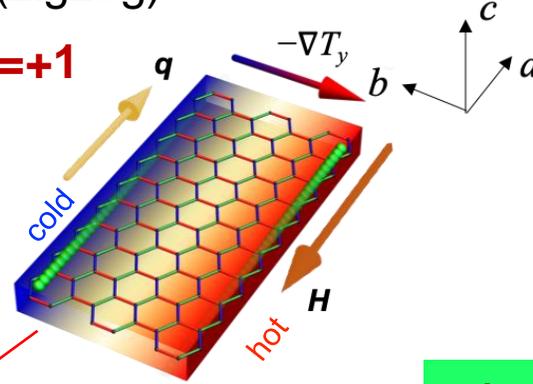
T. Yokoi, YM et al. Science **373**, 568 (2021).



Planar thermal Hall effect

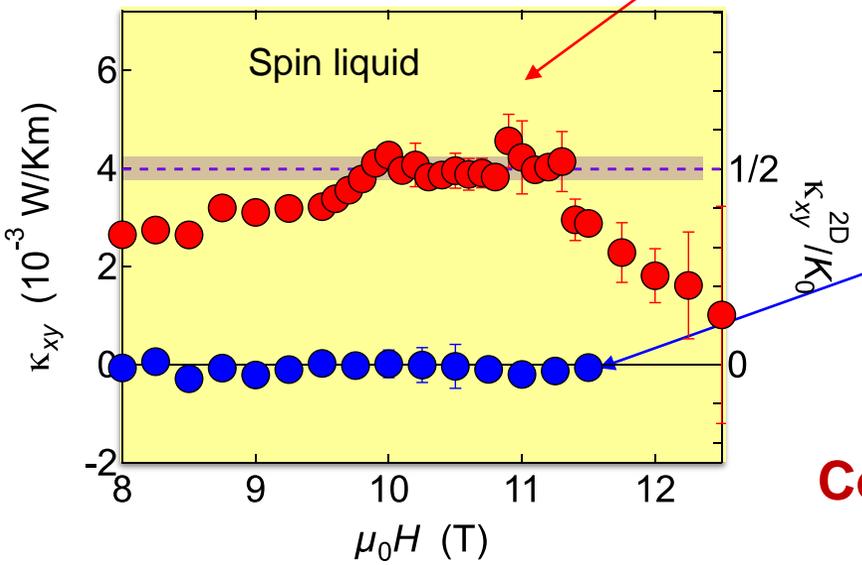
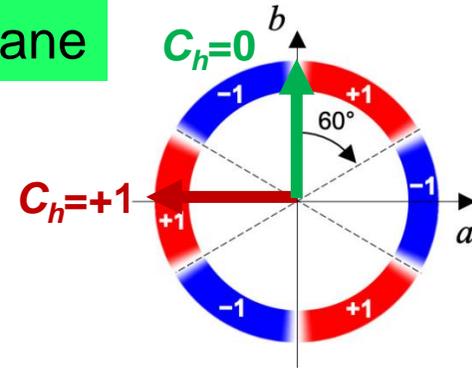
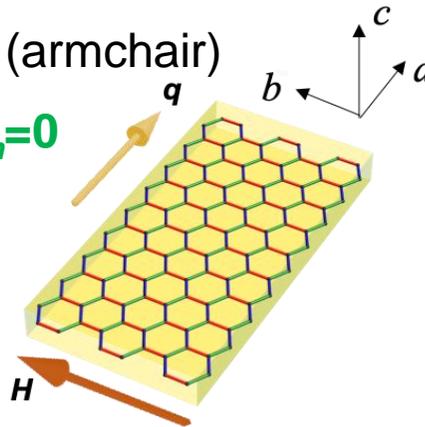
$H // -a$ (zigzag)

$$C_h = +1$$



$H // b$ (armchair)

$$C_h = 0$$

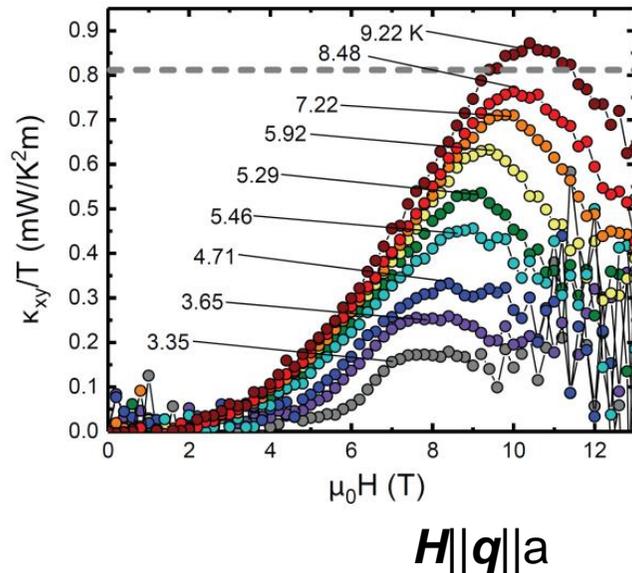


No Majorana edge current

Consistent with Majorana band of Kitaev QSL

Origin of planar thermal Hall: Majorana or magnon ?

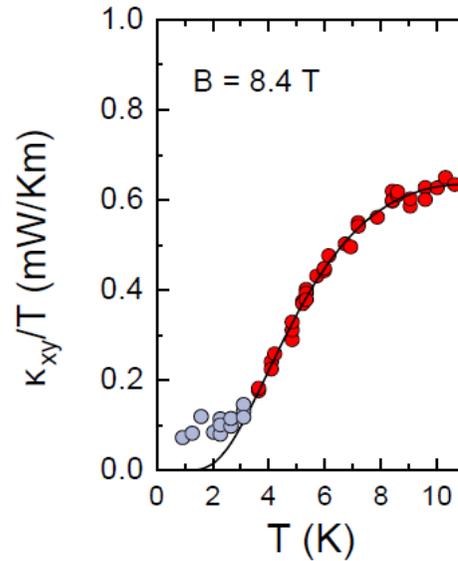
Bosonic scenario



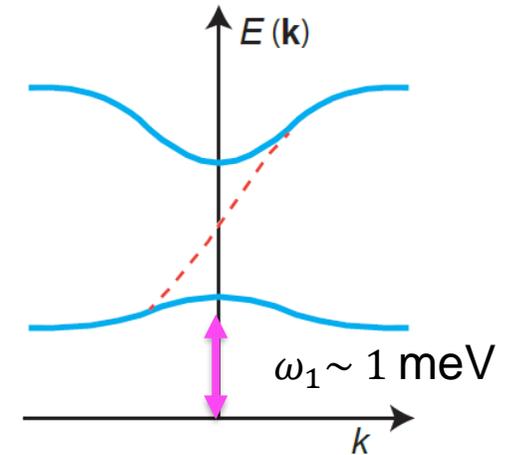
No quantization

P. Czajka *et al.* Nature Phys. **17**, 915 (2021).

P. Czajka *et al.*, Nature Mater. (2022)



κ_{xy}/T decreases with T



bosonic edge excitations
(topological magnons)

Magnon Chern number $\nu \sim 1$

Topological magnons

Sign of κ_{xy} can be explained by topological magnon thermal Hall effect in specific parameter range

E. Z. Zhang *et al.*, Phys Rev B **103**, 174402 (2021).

L. Chern *et al.*, Phys. Rev. Lett. **126**, 147201 (2021).

How to distinguish Majorana and magnon ?

Majorana fermions

Topological magnons
(boson)

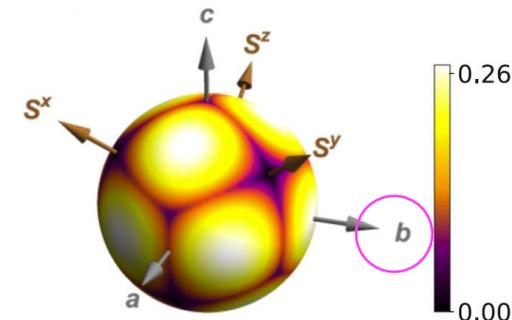
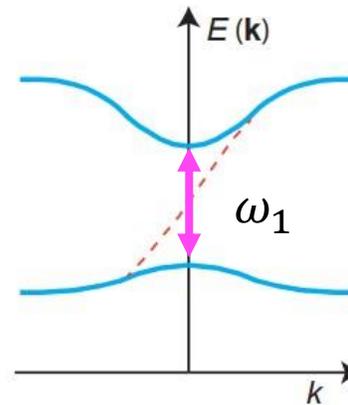
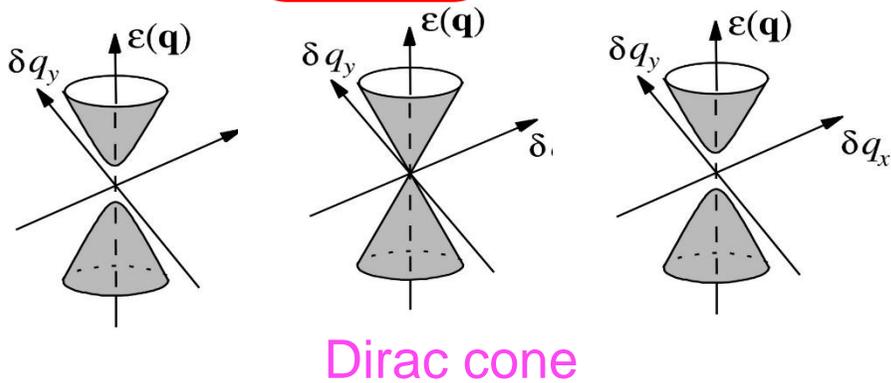
$H \parallel -a$	$H \parallel b$	$H \parallel a$		$H \parallel -a$	$H \parallel b$	$H \parallel a$
+1	0	-1	Chern number	± 1	0	∓ 1
gapped	gapless	gapped	gap	gapped	gapped	gapped

How to distinguish Majorana and magnon ?

Majorana fermions

Topological magnons (boson)

$H \parallel -a$	$H \parallel b$	$H \parallel a$		$H \parallel -a$	$H \parallel b$	$H \parallel a$
+1	0	-1	Chern number	± 1	0	∓ 1
gapped	gapless	gapped	gap	gapped	gapped	gapped



For Majorana, gap closes at $H \parallel b$ \leftrightarrow

For magnon, gap (~ 1 meV) opens at $H \parallel b$ in polarized state

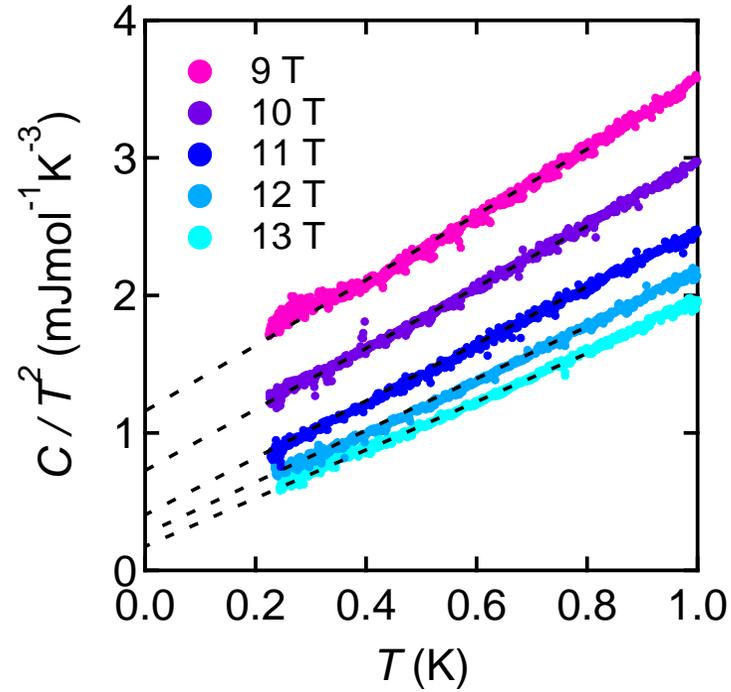
S. Koyama and J. Nasu, Phys. Rev. B **104**, 075121 (2021).

K. Hwang, E-G. Moon *et al.* Nature Commun. **13**, 1 (2021).

Rigorous testing of the Kitaev QSL: **Dirac cone for $H \parallel b$**

Dirac cone for $H \parallel b$: specific heat in FIQD state

$H \parallel b$
armchair



Dirac cone for $H//b$: specific heat in FIQD state

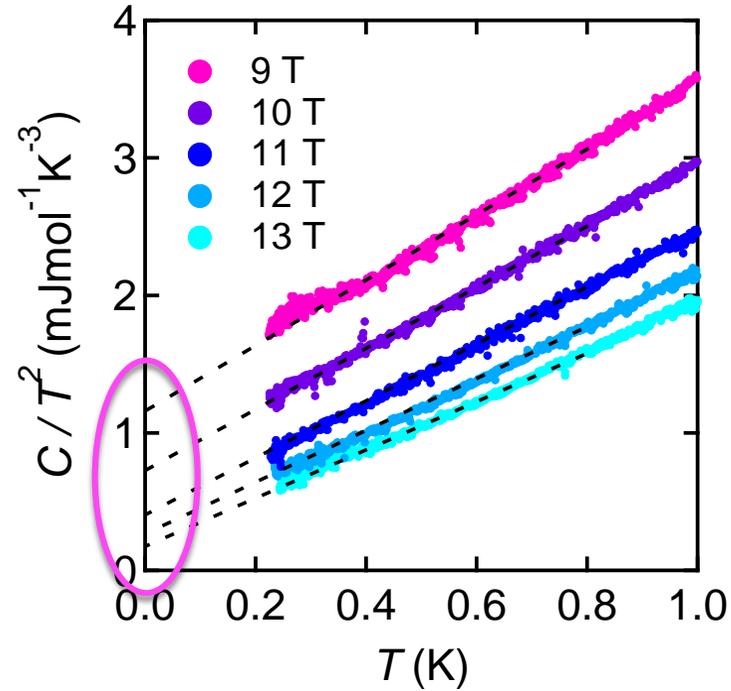
$H \parallel b$
armchair



Finite intercepts at $T \rightarrow 0$

$$C \propto T^2$$

over a wide H range
above $H_{AF} \sim 7T$



Dirac cone for $H//b$: specific heat in FIQD state

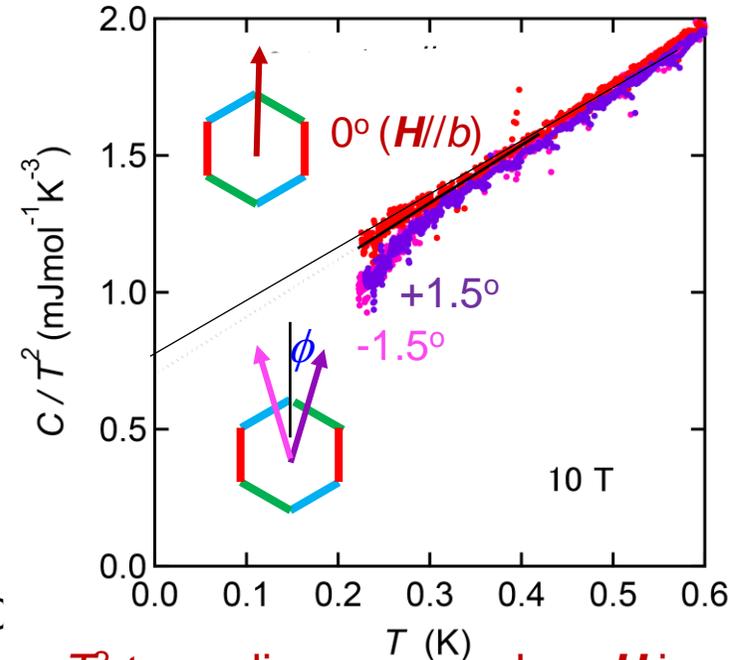
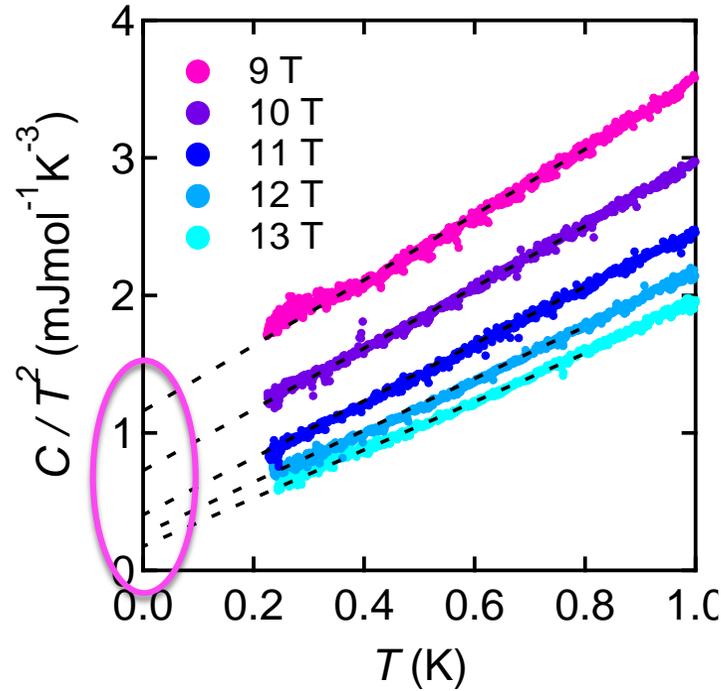
$H \parallel b$
armchair



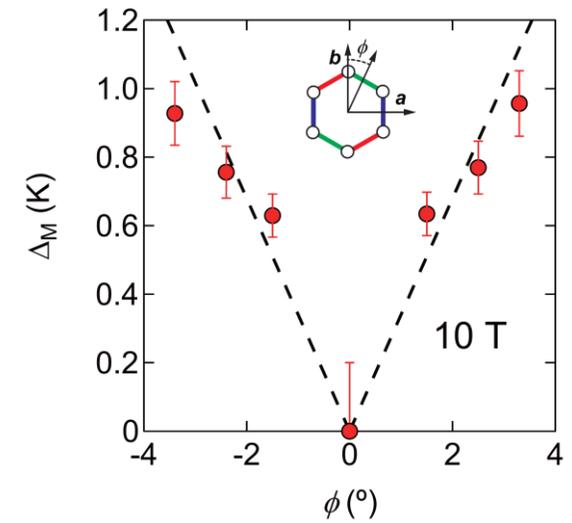
Finite intercepts at $T \rightarrow 0$

$$C \propto T^2$$

over a wide H range
above $H_{AF} \sim 7T$

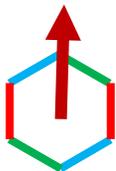


T^2 -term disappears when H is
slightly away from b -axis



Dirac cone for $H//b$: specific heat in FIQD state

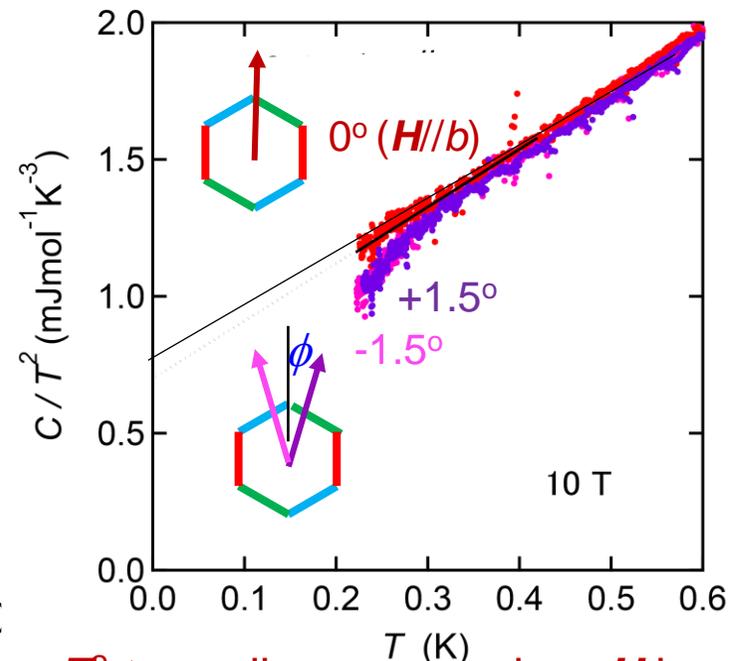
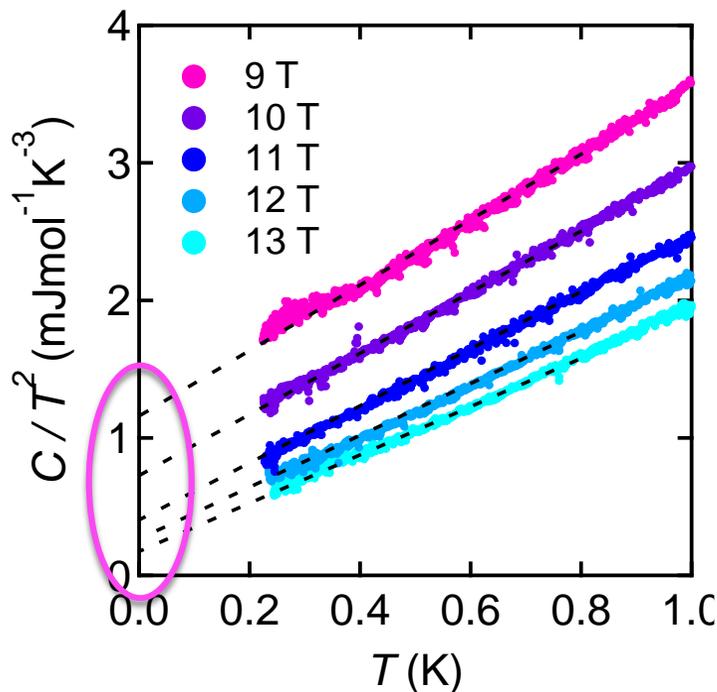
$H // b$
armchair



Finite intercepts at $T \rightarrow 0$

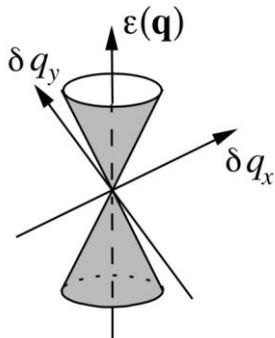
$$C \propto T^2$$

over a wide H range
above $H_{AF} \sim 7T$



T^2 -term disappears when H is slightly away from b -axis

Incompatible with 2D
magnon and phonon

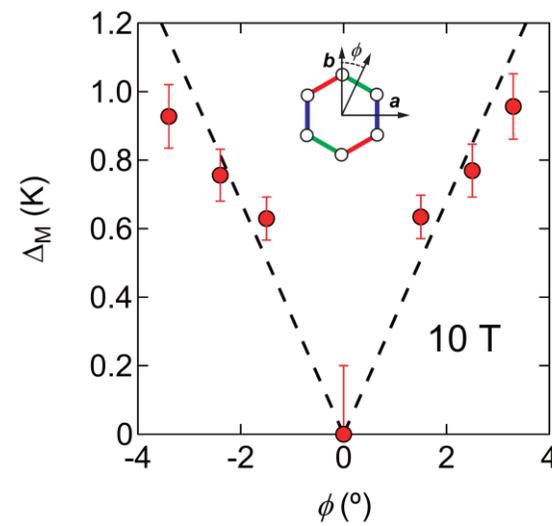


$$C_{\text{mag}}/T \propto T$$

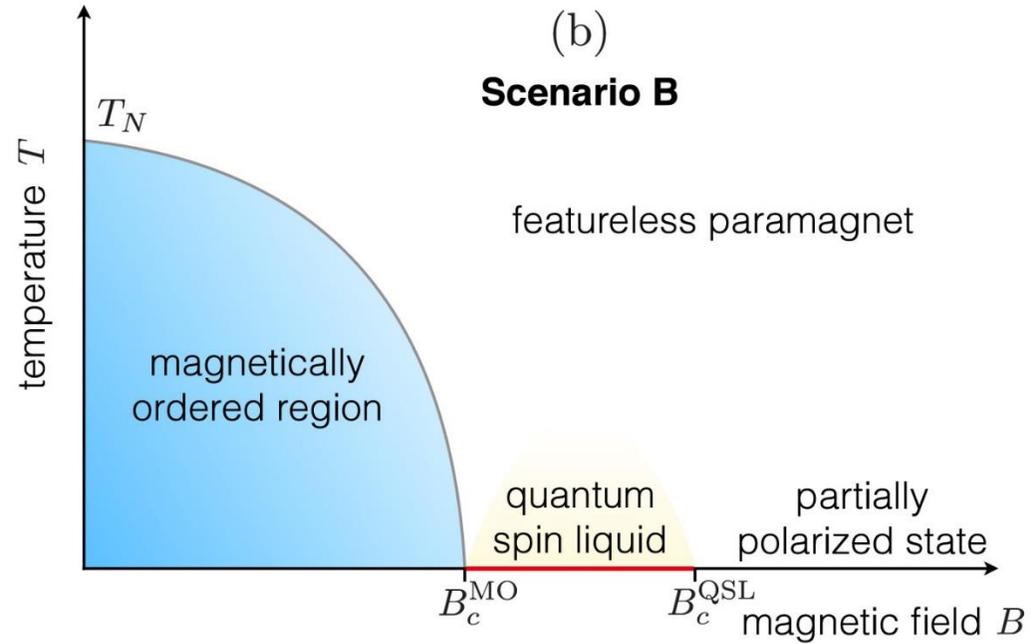
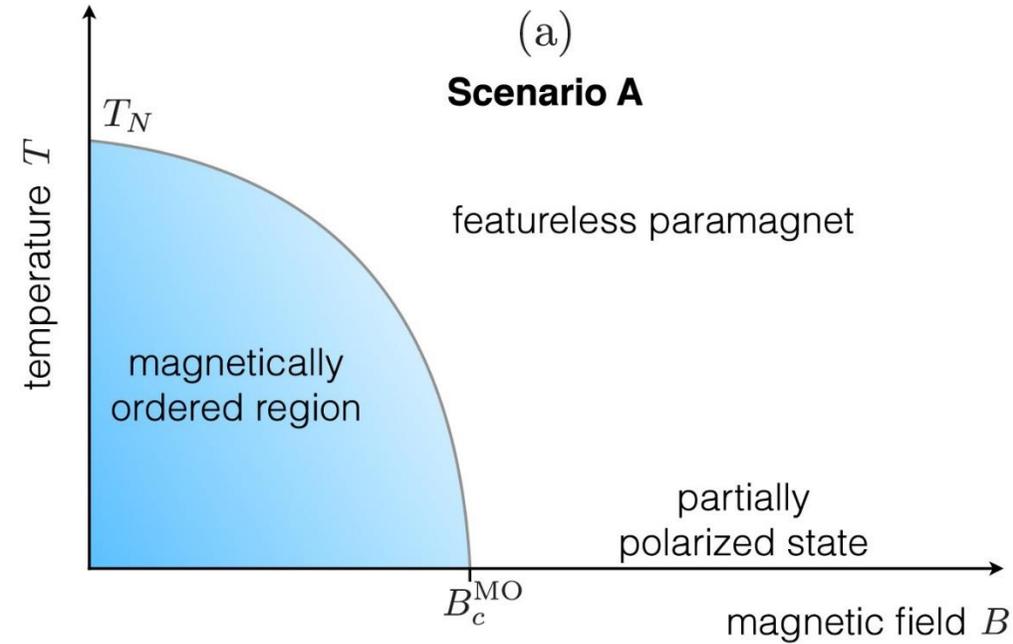
Linear dispersion

Dirac cone

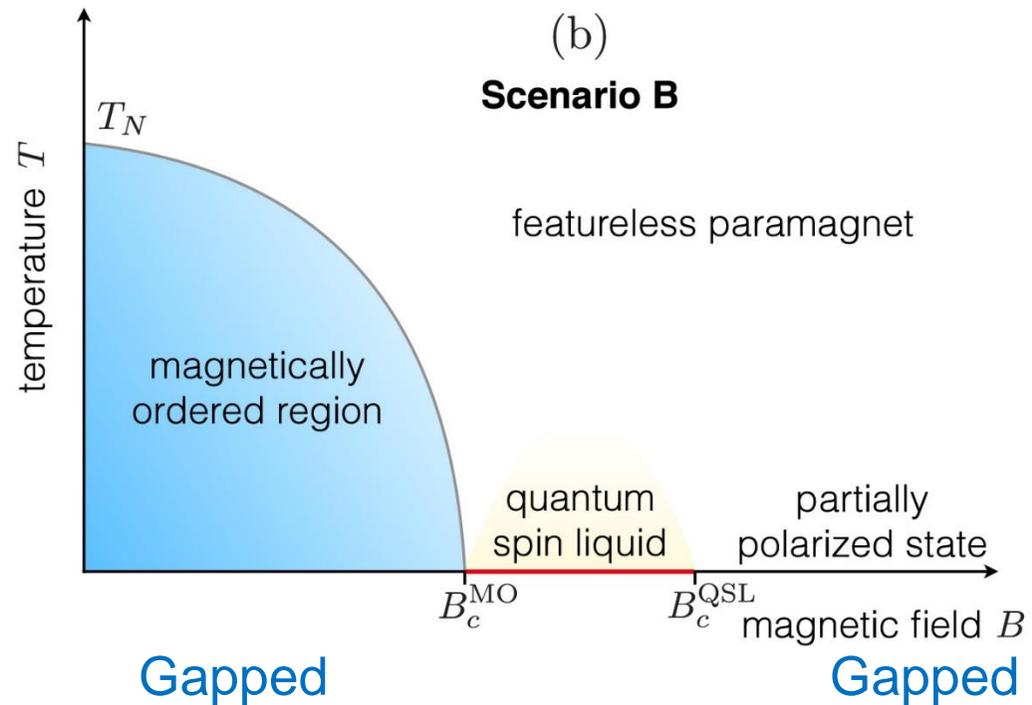
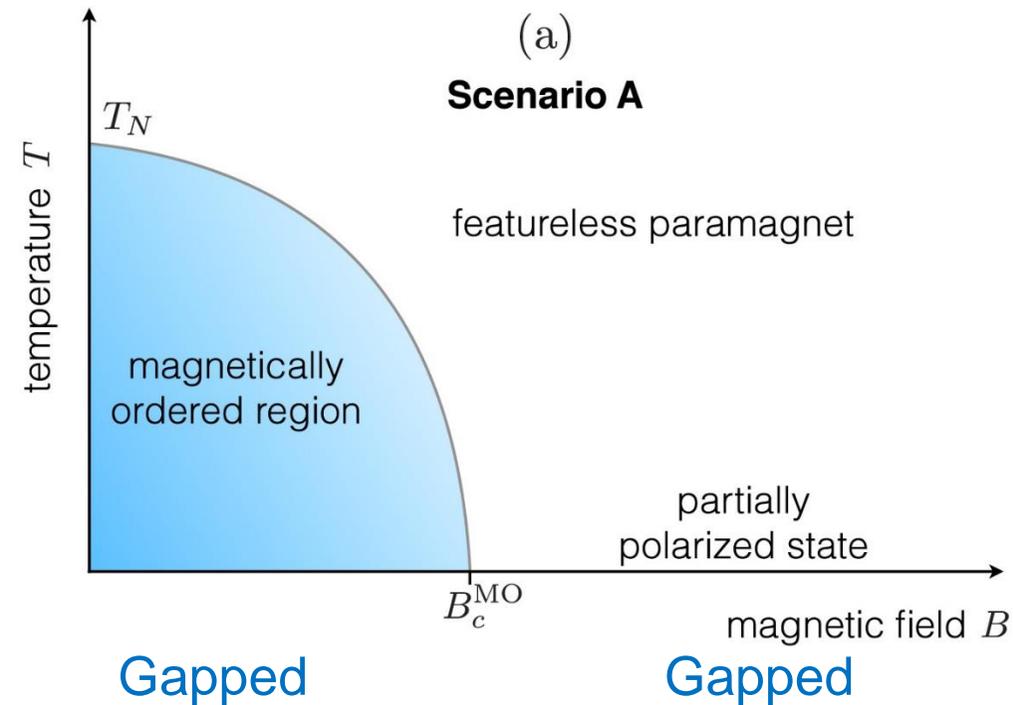
Consistent with the Kitaev QSL



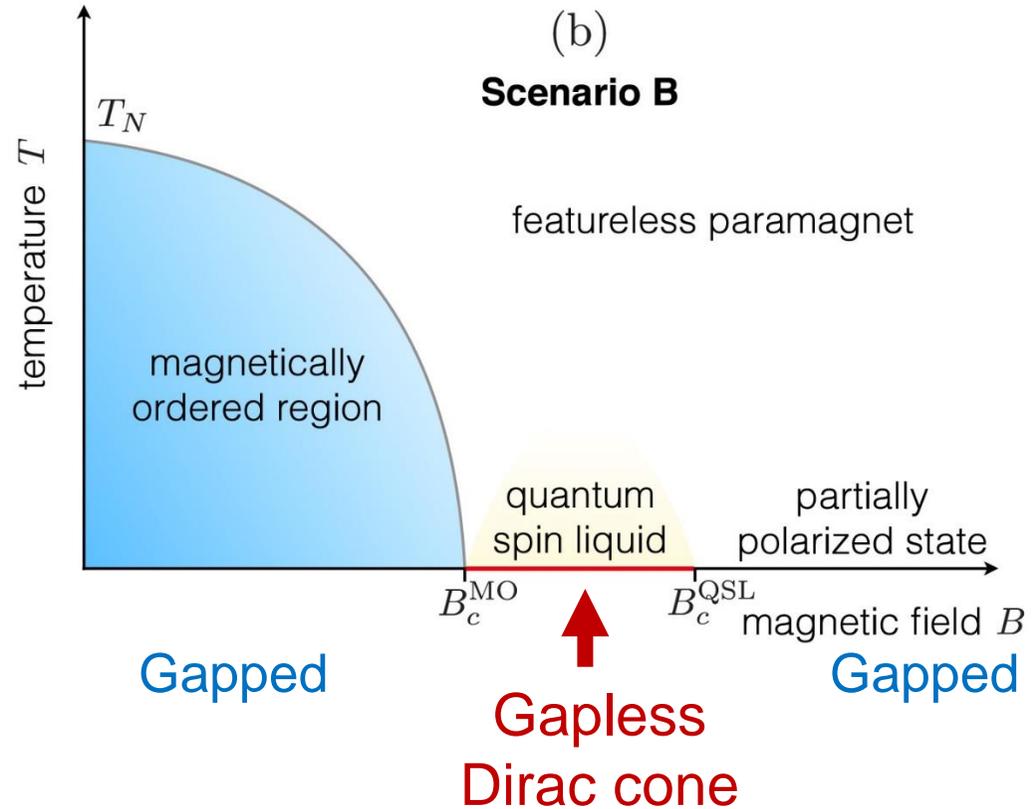
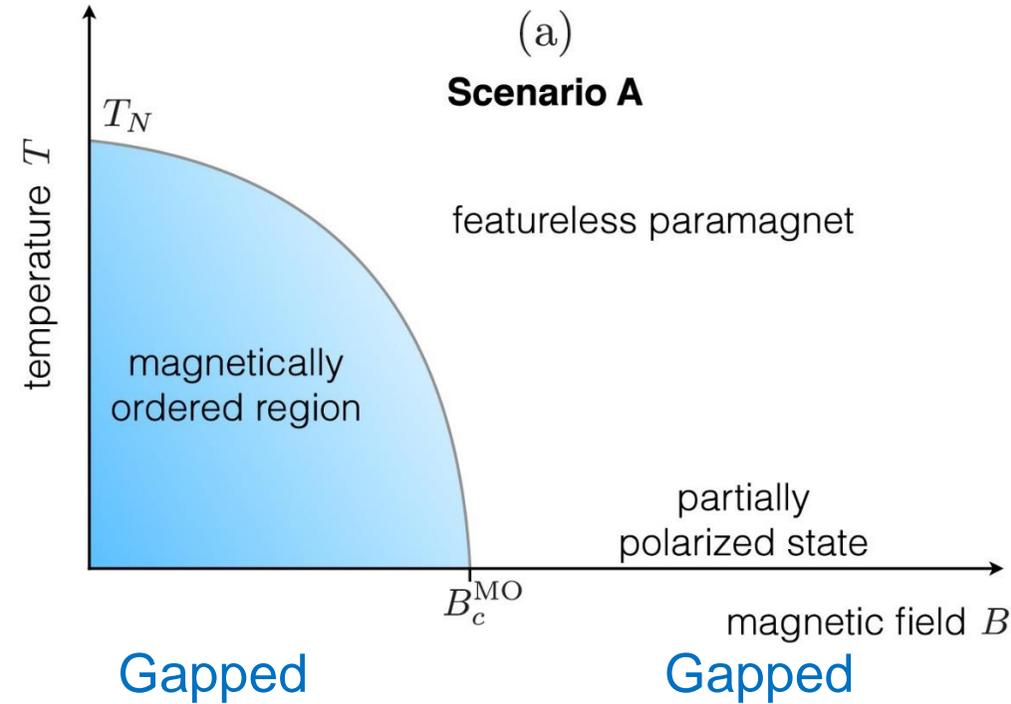
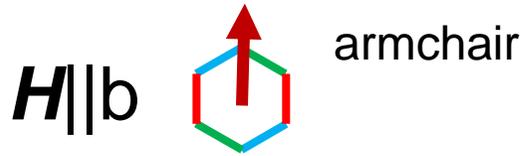
Phase diagram



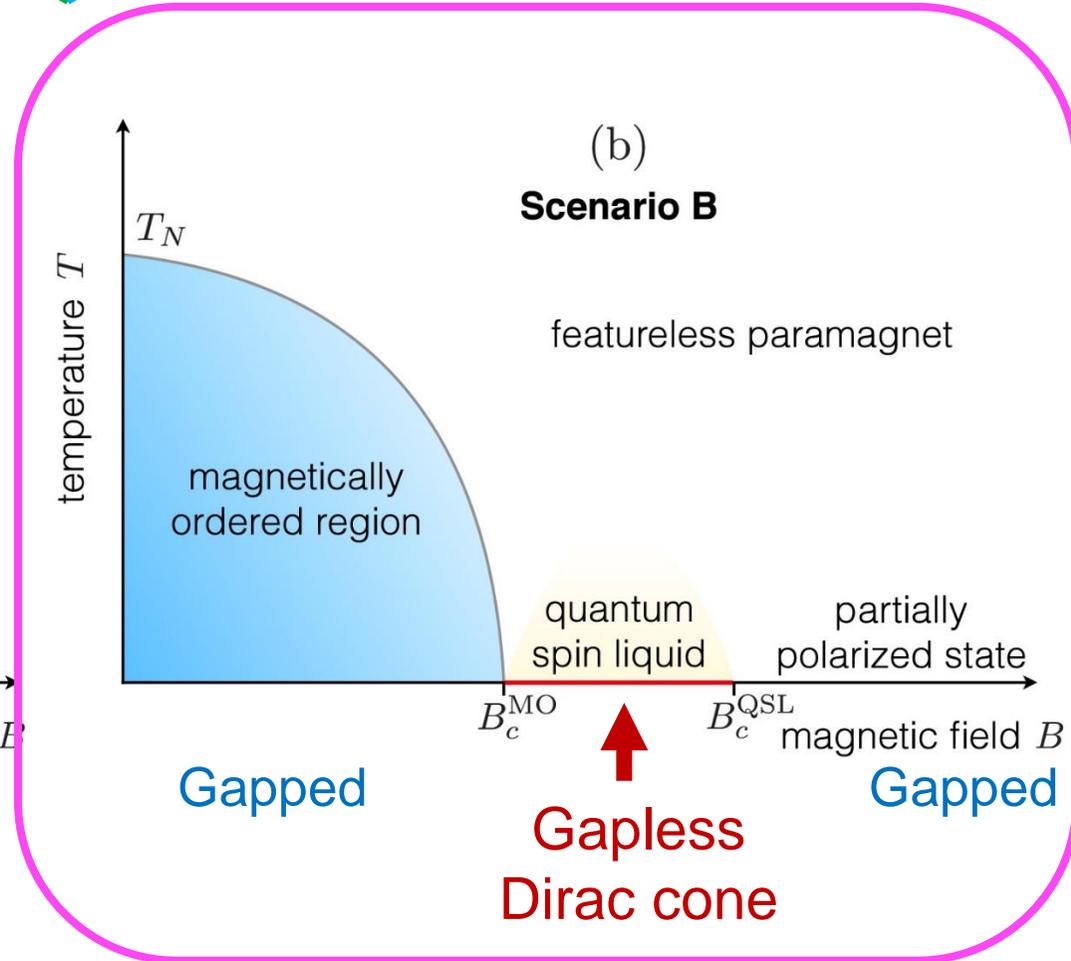
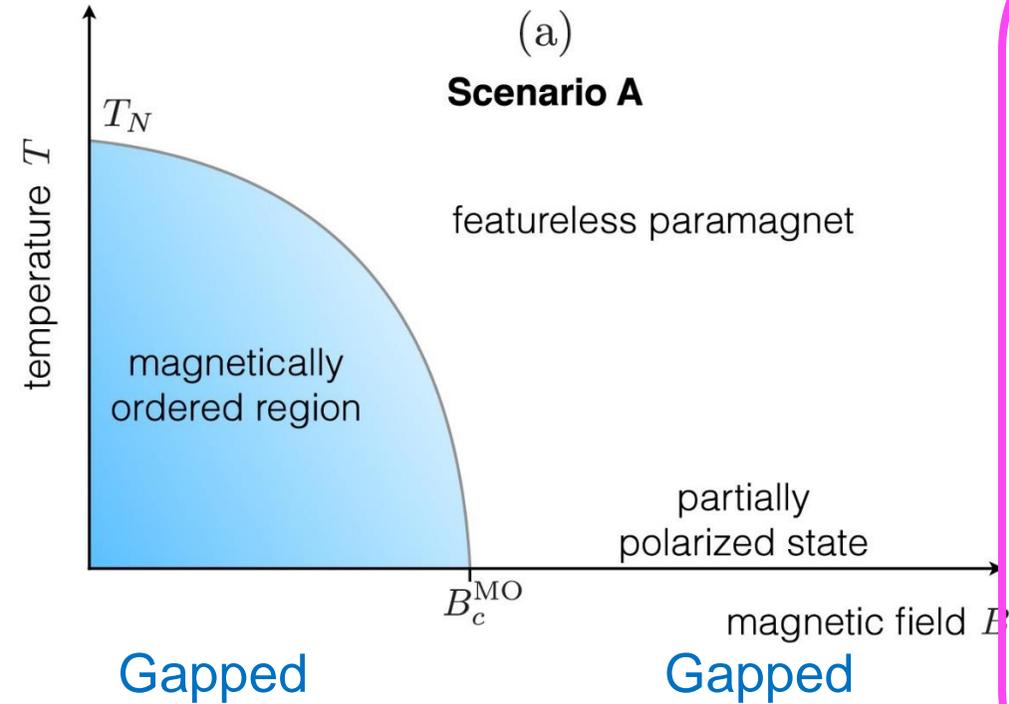
Phase diagram



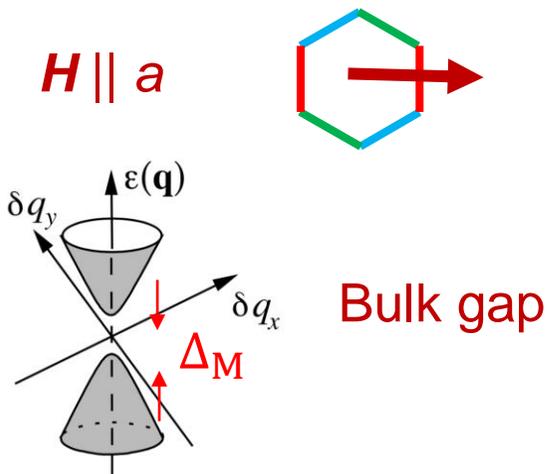
Phase diagram



Phase diagram



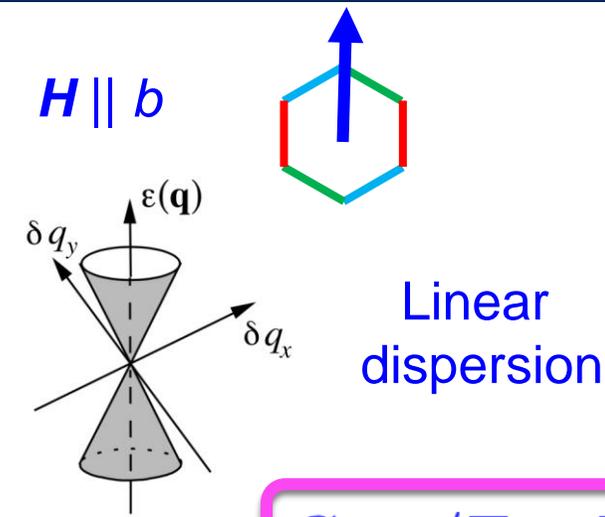
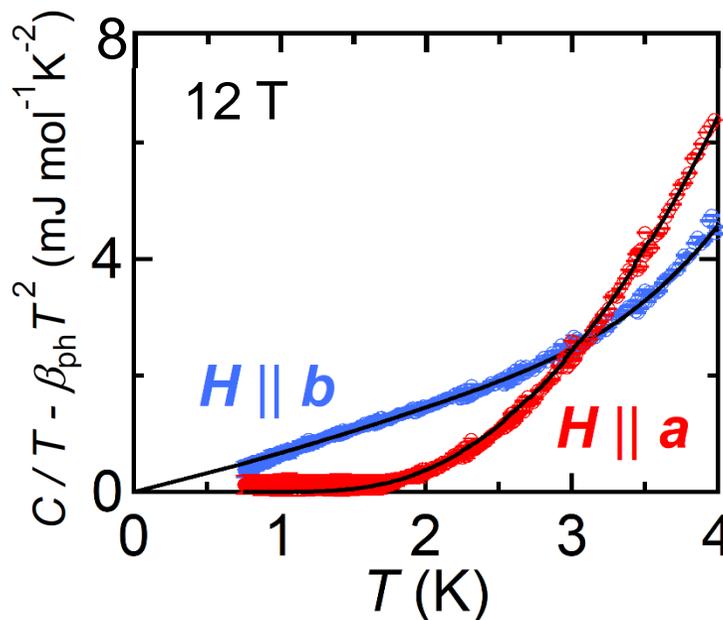
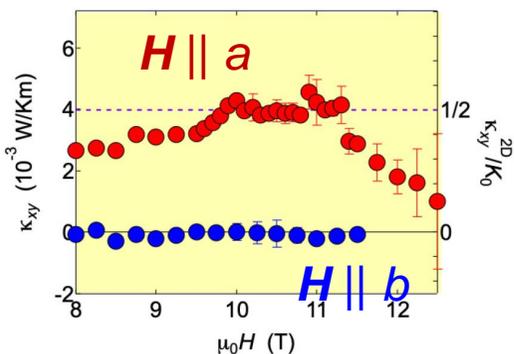
Majorana gap and Dirac cone in FIQD state



$$C_{\text{mag}}/T \propto e^{-\frac{\Delta_M}{T}}$$

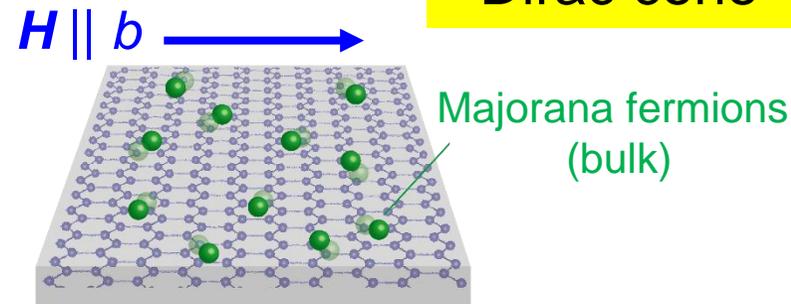
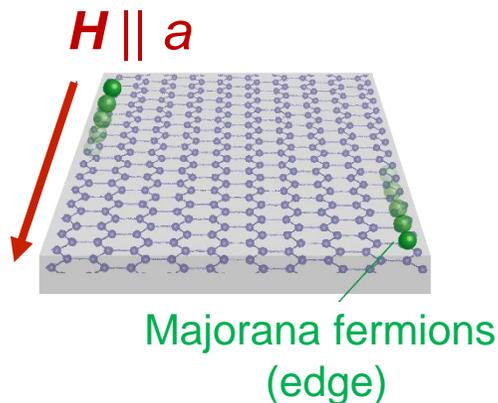
$$\Delta_M \propto \frac{H^3}{\Delta_{\text{flux}}^2}$$

Majorana gap



$$C_{\text{mag}}/T \propto T$$

Dirac cone



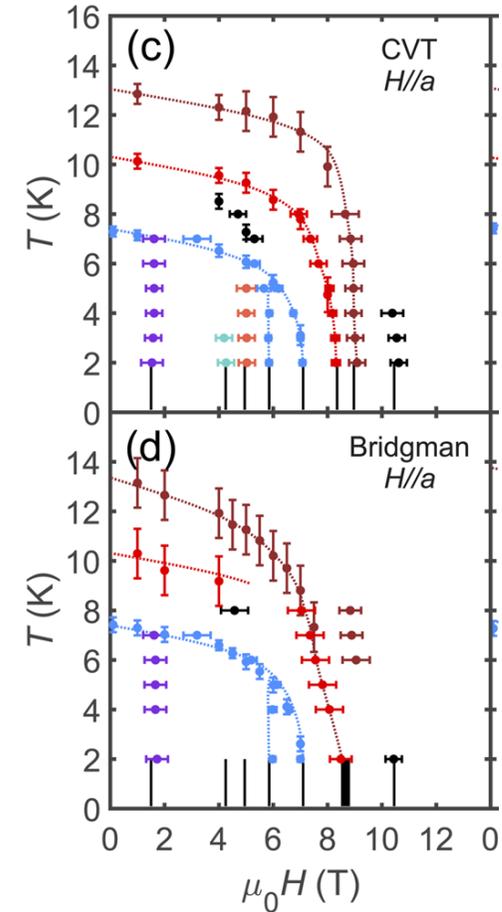
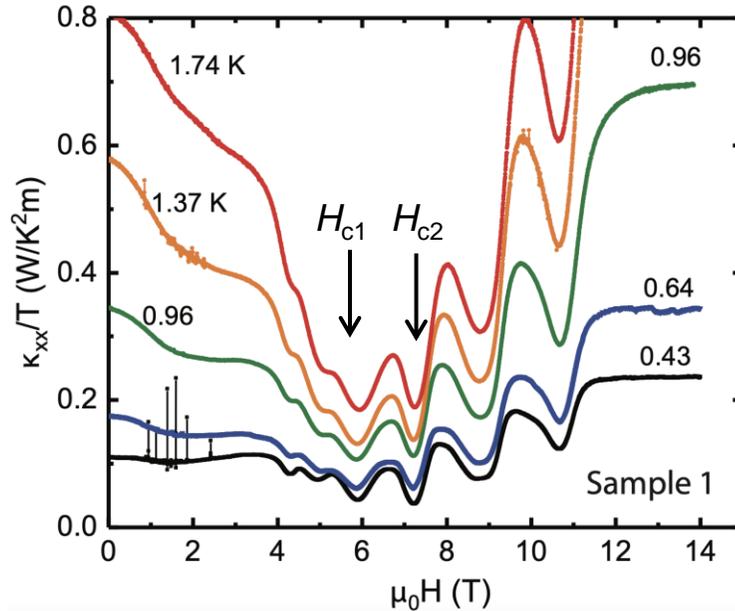
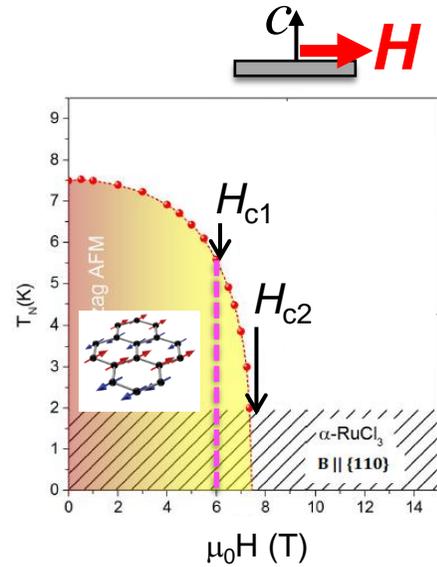
Bulk-edge correspondence

O. Tanaka, YM, T. Shibauchi *et al.* Nature Phys. **18**, 429 (2022).

A.U.B. Wolter and C. Hess, Nature Phys. **18**, 378 (2022).

Oscillating thermal conductivity

CVT



Quantum oscillations of fermionic quasiparticles

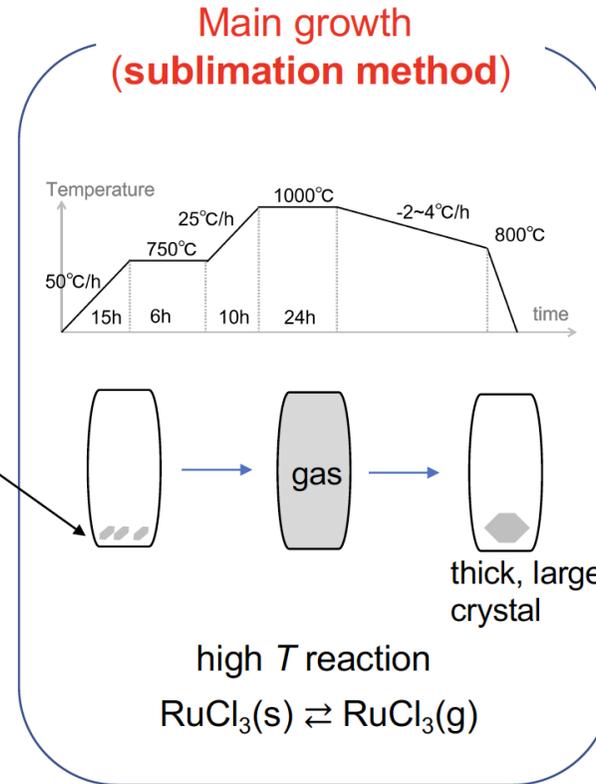
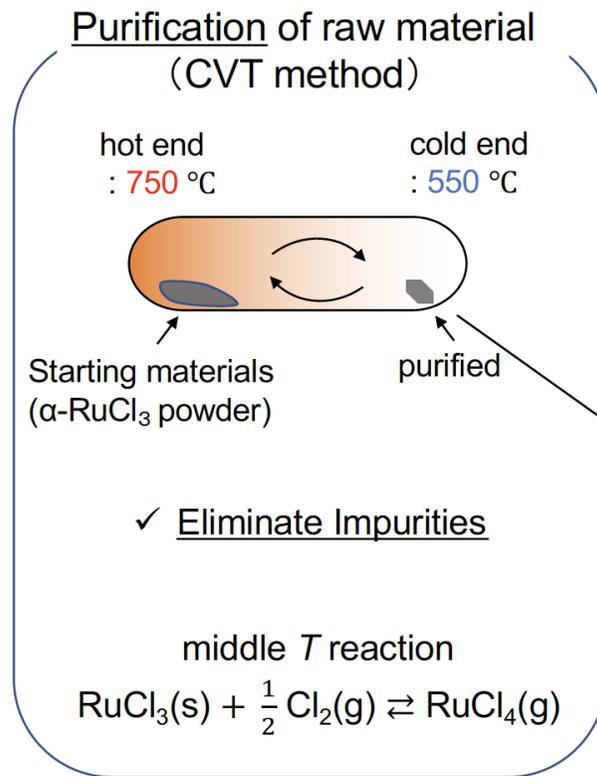
P. Czajka *et al.* Nature Phys. **17**, 915 (2021).

Magnetic transitions due to stacking faults

J.A.N. Bruin *et al.* APL Mater. **10**, 090703 (2022).

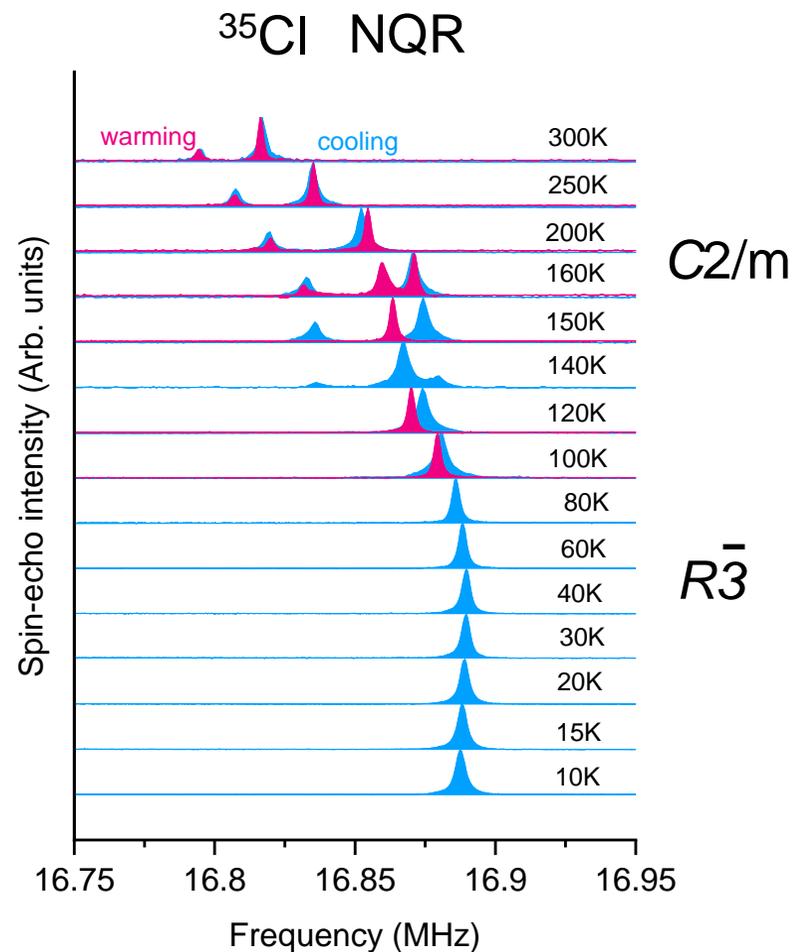
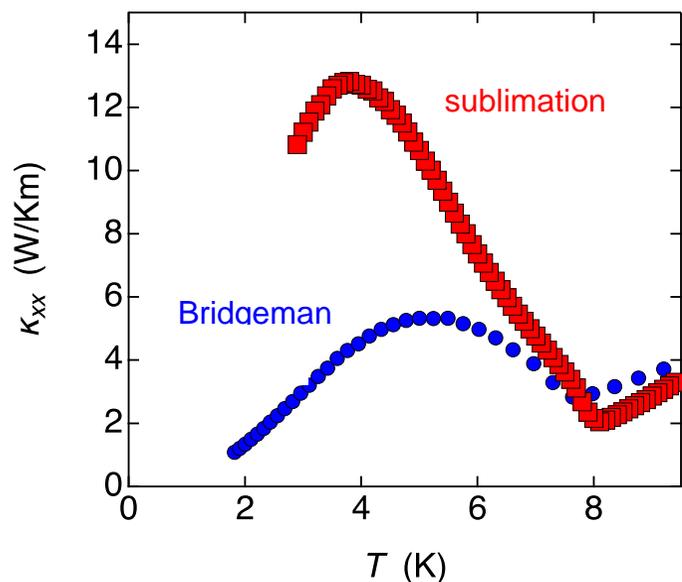
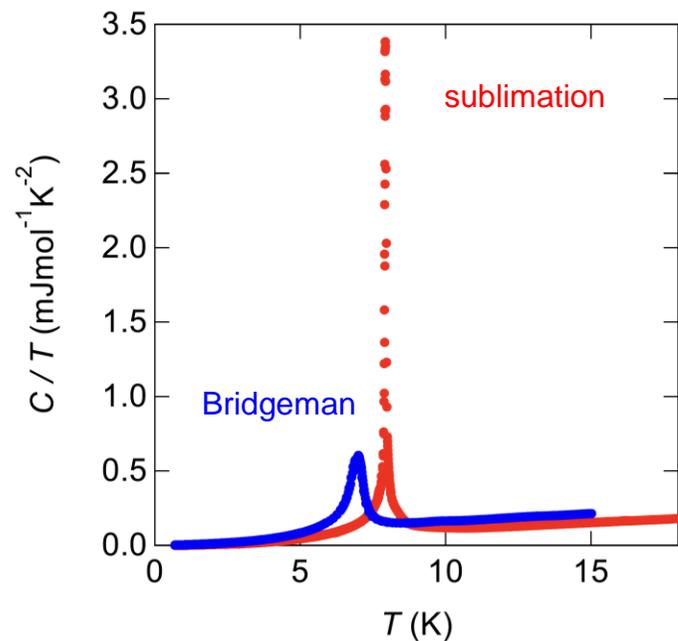
E. Lefrancois *et al.* PRB **107**, 064408 (2023).

Very clean α -RuCl₃ single crystals



Disorders and defects can
be removed by slow growth

Very clean α -RuCl₃ single crystals



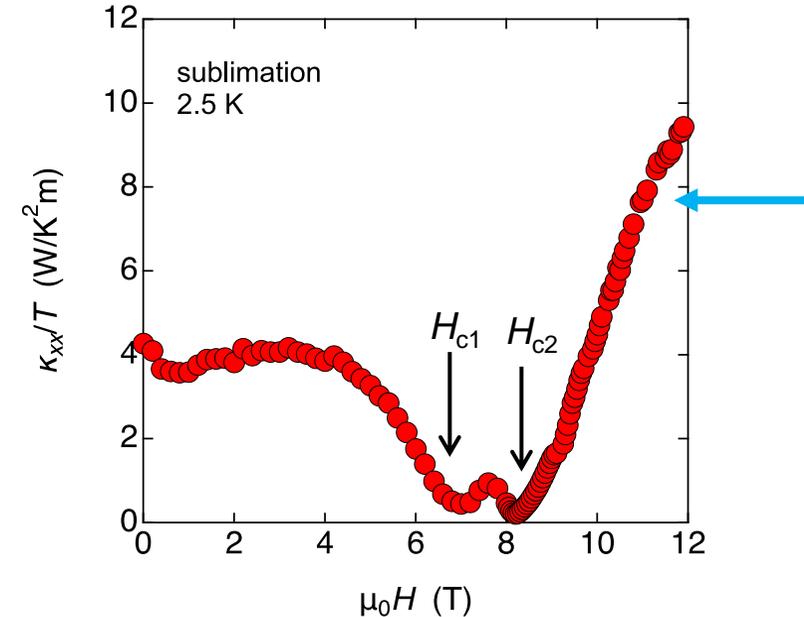
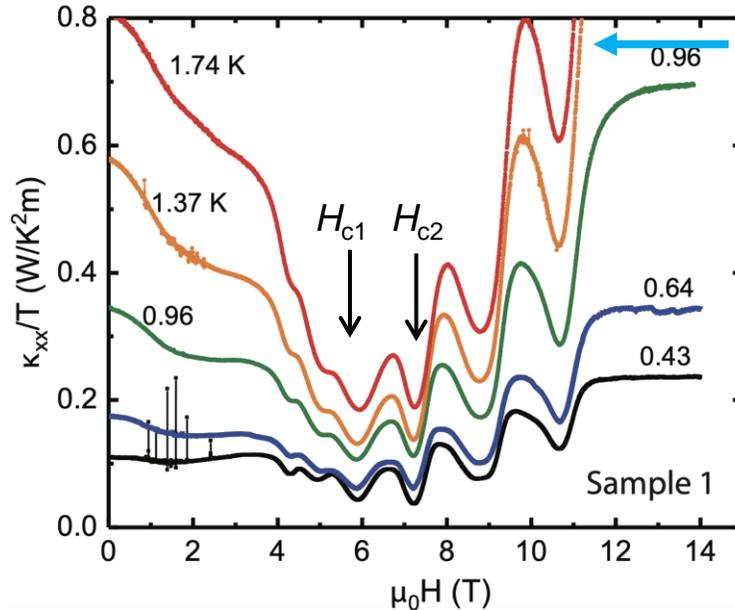
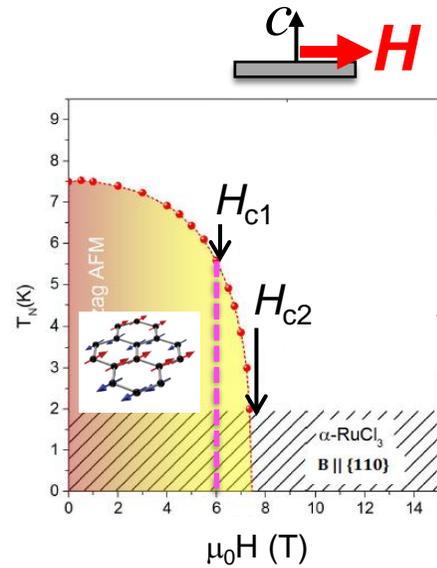
Y. Shimizu *et al.* unpublished

R. Namba, YM, T. Shibauchi *et al.* in preparation.

Oscillating thermal conductivity

CVT

Sublimation



Quantum oscillations of fermionic quasiparticles

P. Czajka *et al.* Nature Phys. **17**, 915 (2021).

No oscillations in very clean crystals

Y. Xing, YM *et al.* in preparation.

Magnetic transitions due to stacking faults

J.A.N. Bruin *et al.* APL Mater. **10**, 090703 (2022).

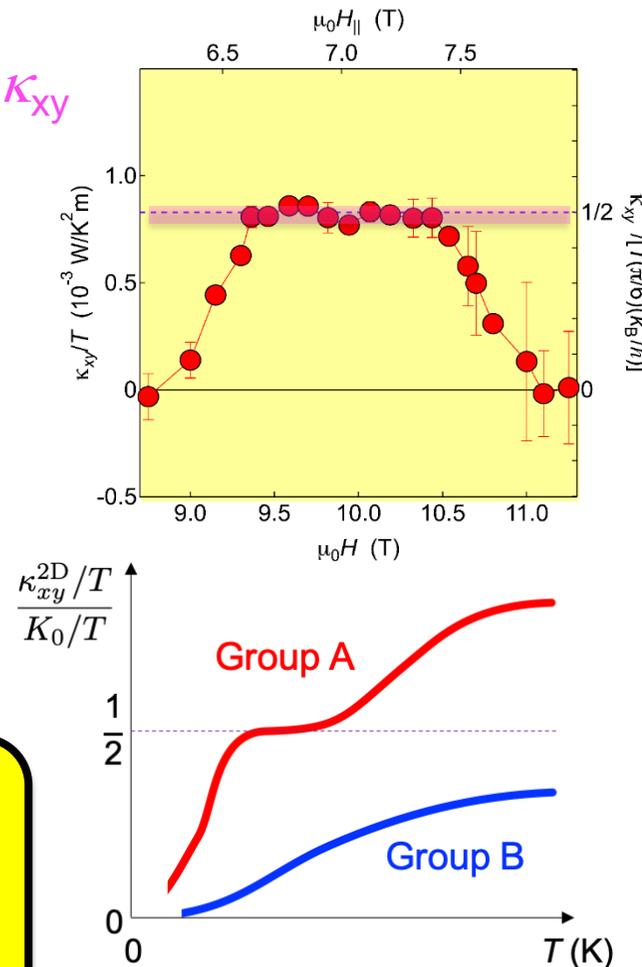
E. Lefrancois *et al.* PRB **107**, 064408 (2023).

Origin of the thermal Hall effect

Some people are not still convinced by the quantized κ_{xy}

1. We cannot determine κ_{xy} accurately
2. The deviation from the quantized value at low temperatures
3. We cannot fully explain the sample dependent quantization.

We need a new approach to tackle the quantized thermal Hall issue **without relying on the MAGNITUDE of κ_{xy}** .



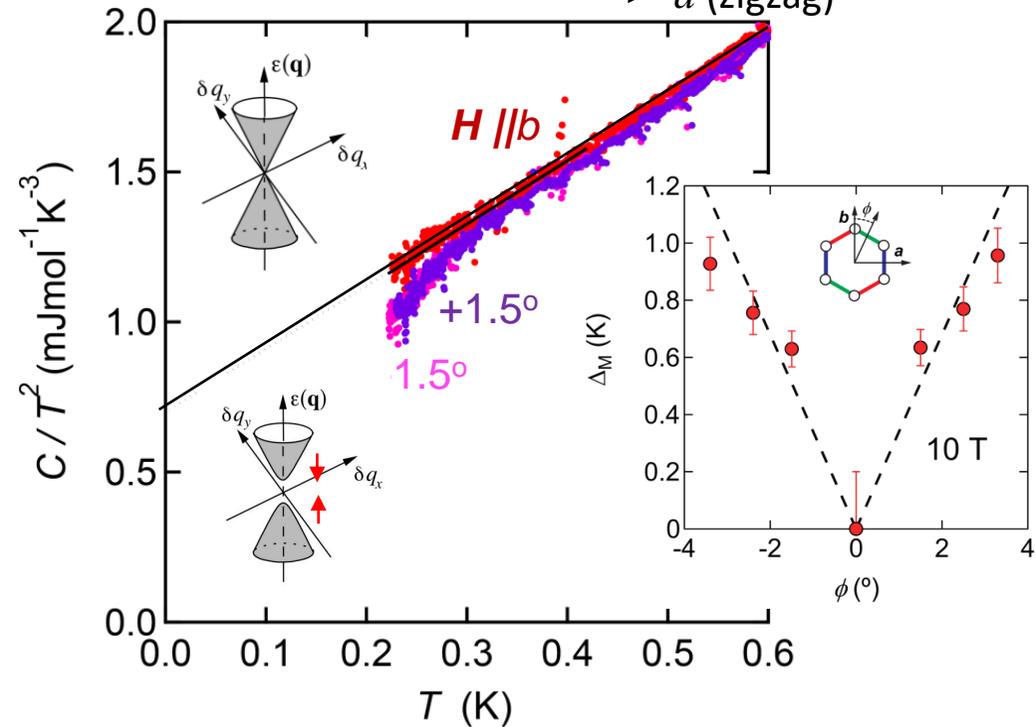
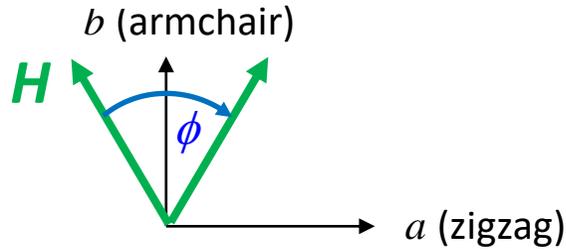
Origin of planar thermal Hall: bosonic or fermionic ?

In-plane H rotation across b -axis

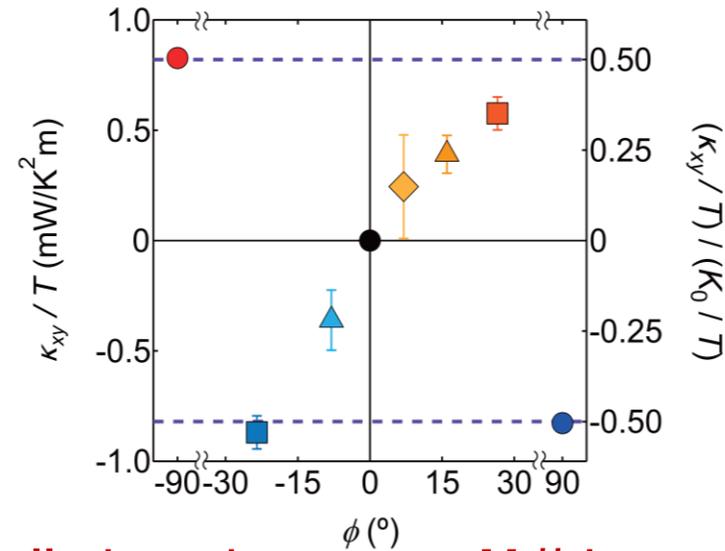
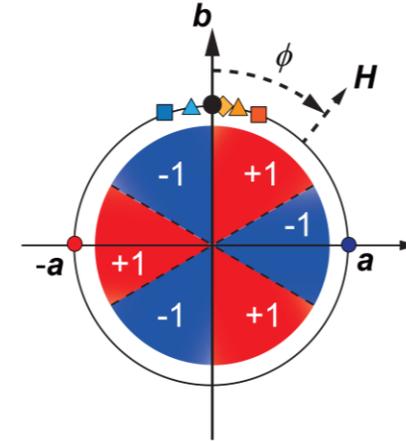
K. Imamura, S. Suetsugu *et al.* arXiv:2305.10619.

① Specific heat

② Planar thermal Hall effect



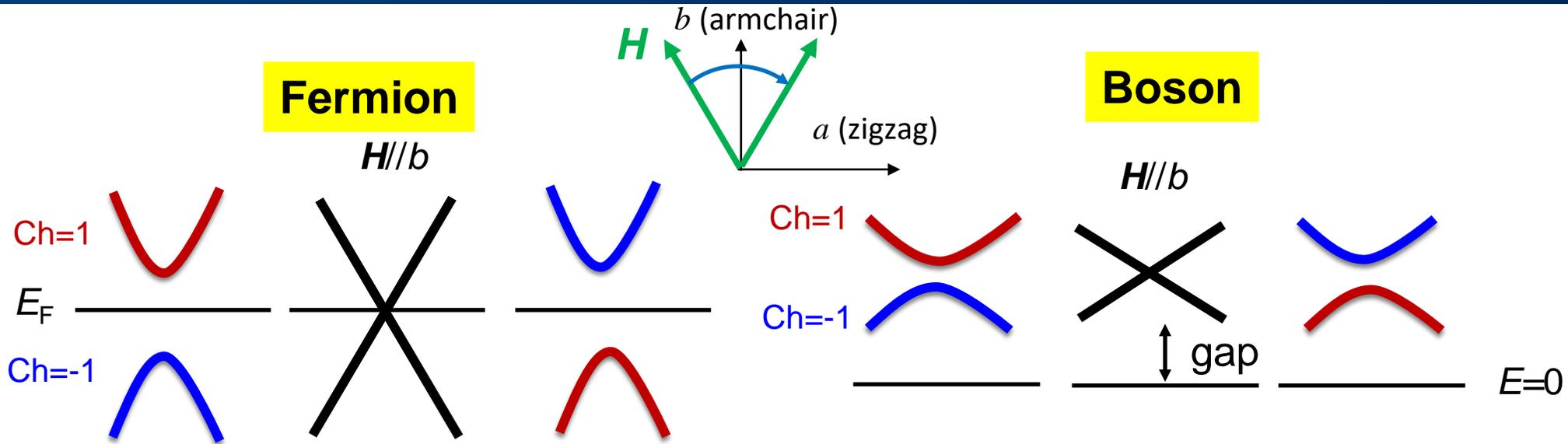
Gap closing at $H \parallel b$



Hall sign change at $H \parallel b$

Two distinct features are observed concurrently at $H \parallel b$

Origin of planar thermal Hall: bosonic or fermionic ?



Sign change of κ_{xy} is always accompanied by gap closing

Gap is always finite.
Sign change is not related to the gap closing.

In α - RuCl_3

Gap closing

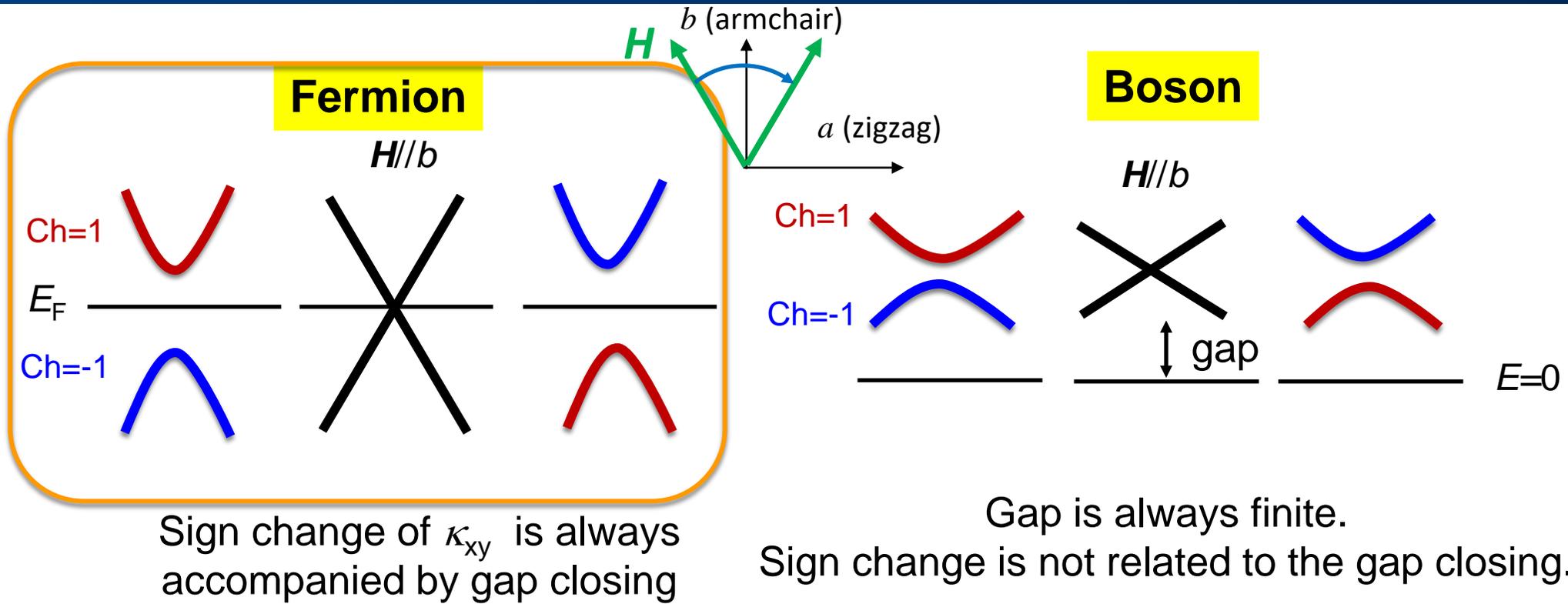
Sign change of the planar thermal Hall effect

Concurrently occur
at $H // b$

Evidence for **fermionic origin** of the thermal Hall effect

In this case, **thermal Hall conductance is quantized.**

Origin of planar thermal Hall: bosonic or fermionic ?



In α -RuCl₃

Gap closing

Sign change of the planar thermal Hall effect

Concurrently occur
 at $H // b$

Evidence for **fermionic origin** of the thermal Hall effect

In this case, **thermal Hall conductance is quantized.**

1. Half integer quantized thermal Hall effect in α - RuCl_3

2. Is α - RuCl_3 really Kitaev material?

Topology: Topological invariant

Thermodynamics: Dirac cone and Majorana gap

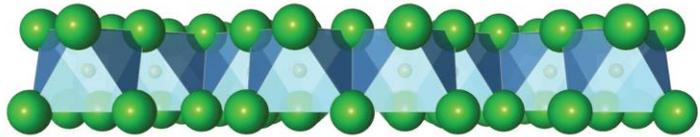
Thermal transport properties of very clean crystals

3. **STM studies on monolayer α - RuCl_3**

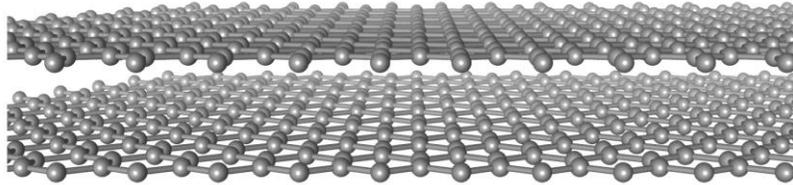
Local density of states around defects

STM image of monolayer α - RuCl_3 on HOPG

Y. Kohsaka, YM, *et al.* a preprint



α - RuCl_3
(monolayer)

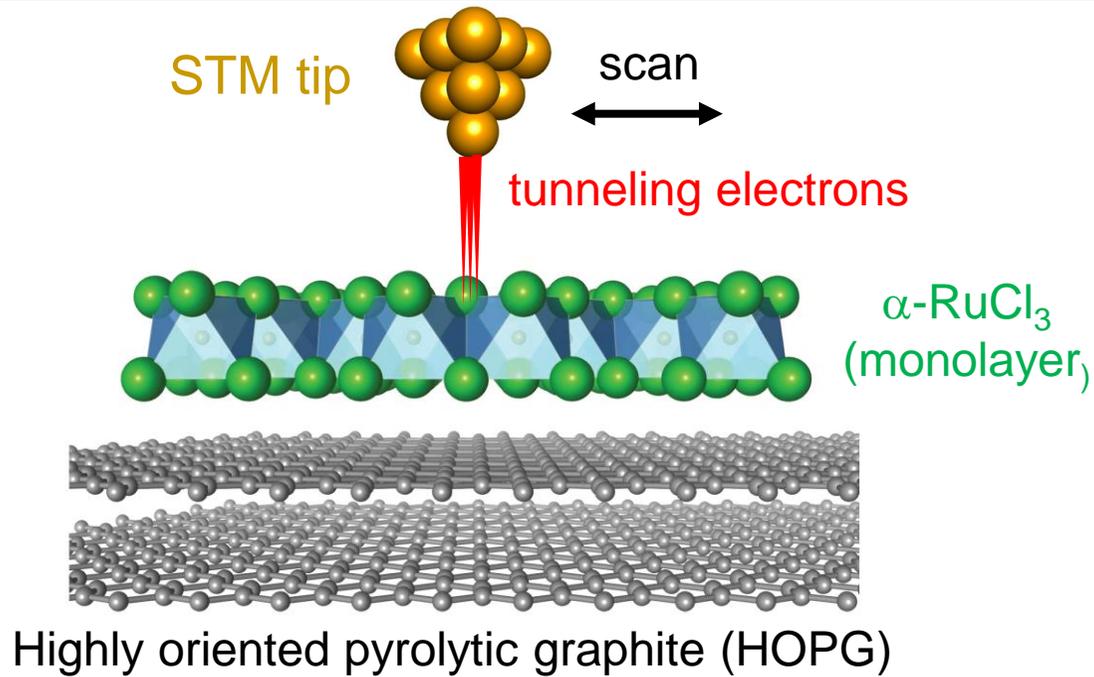


Highly oriented pyrolytic graphite (HOPG)

Ru

STM image of monolayer α -RuCl₃ on HOPG

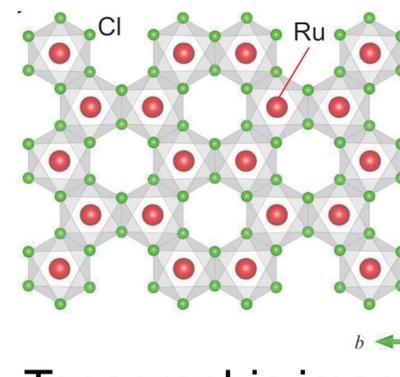
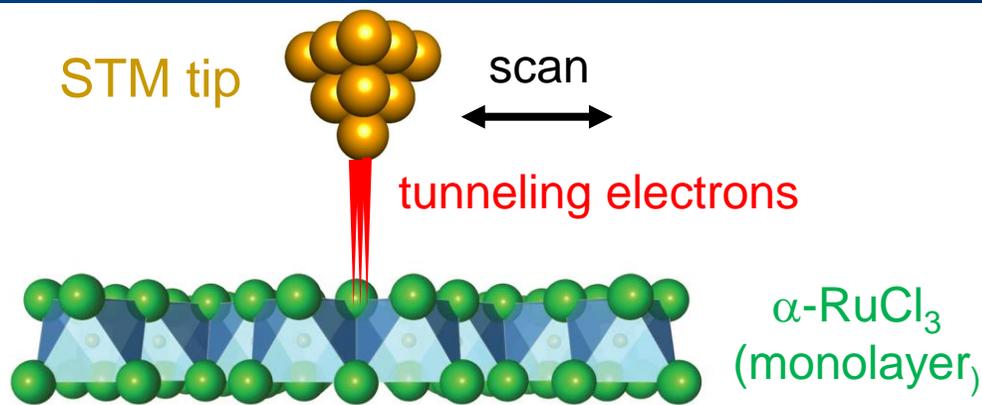
Y. Kohsaka, YM, *et al.* a preprint



Ru

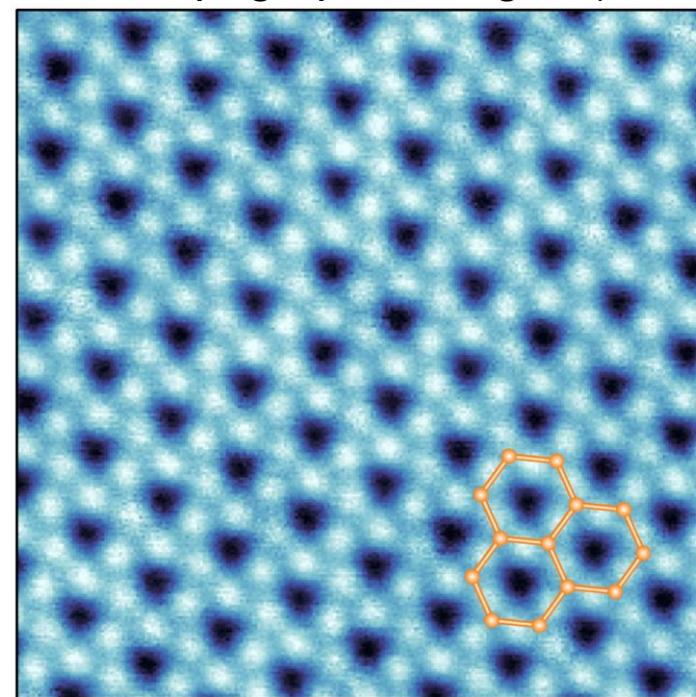
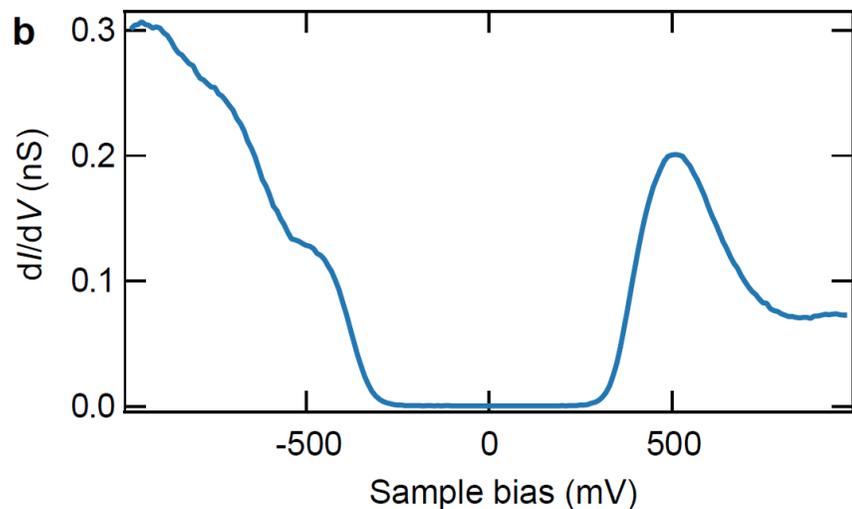
STM image of monolayer α - RuCl_3 on HOPG

Y. Kohsaka, YM, *et al.* a preprint



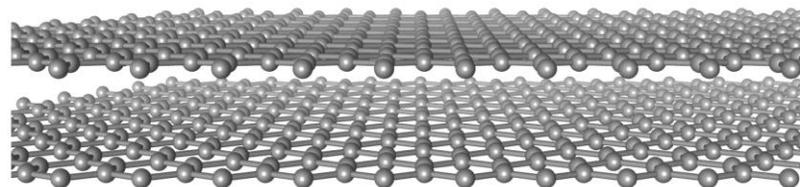
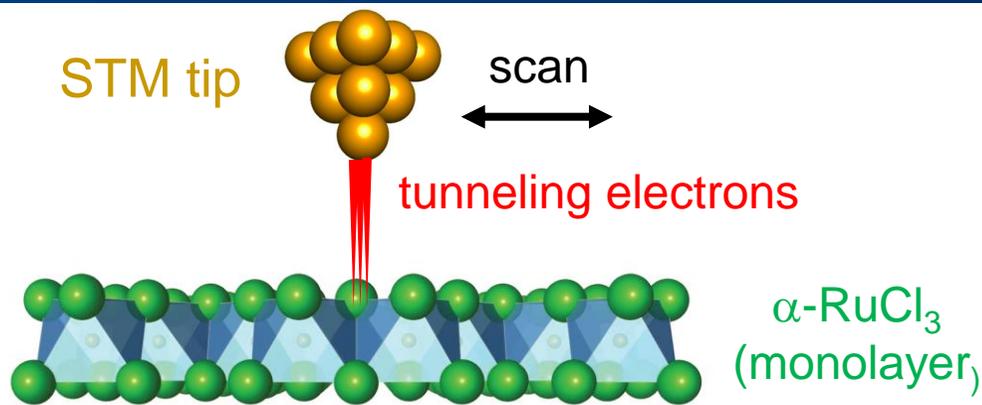
Topographic image (5 K)

Highly oriented pyrolytic graphite (HOPG)

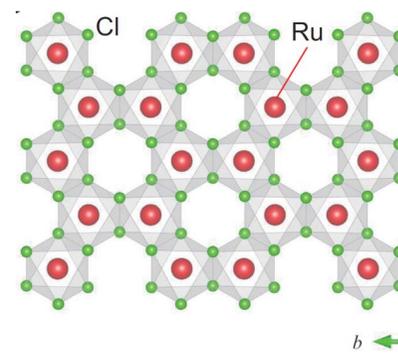


STM image of monolayer α - RuCl_3 on HOPG

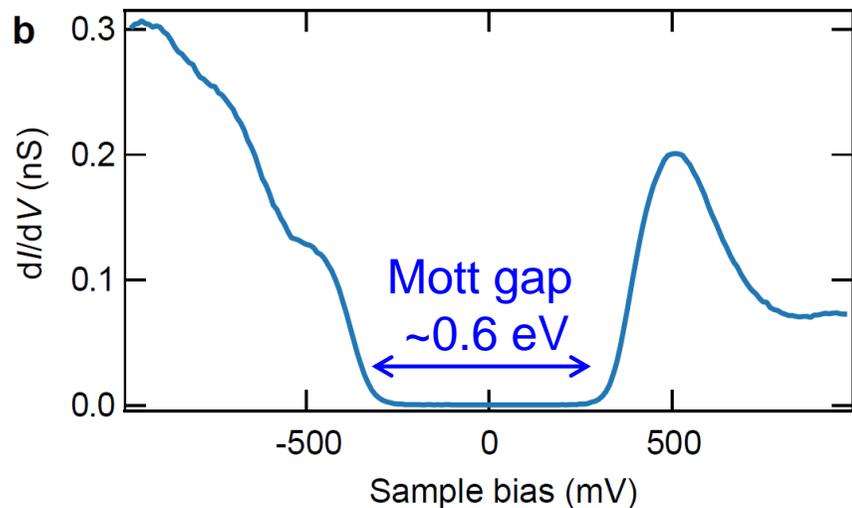
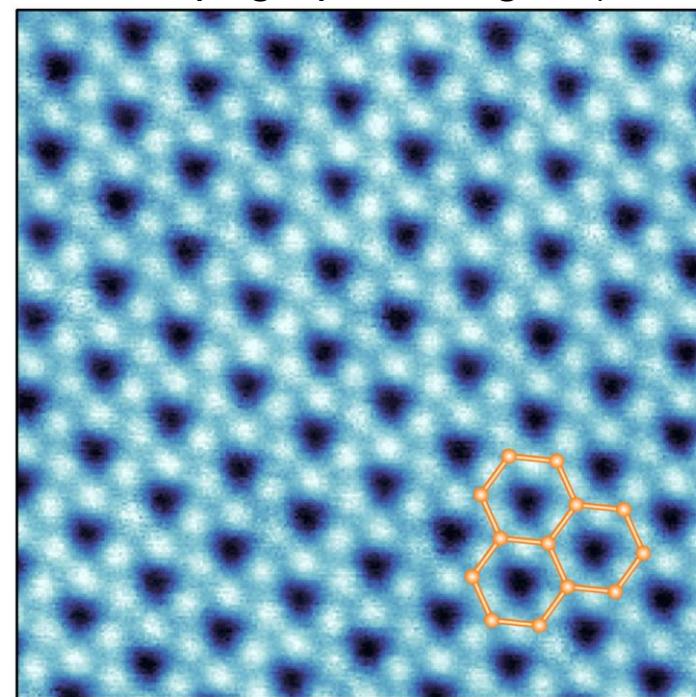
Y. Kohsaka, YM, *et al.* a preprint



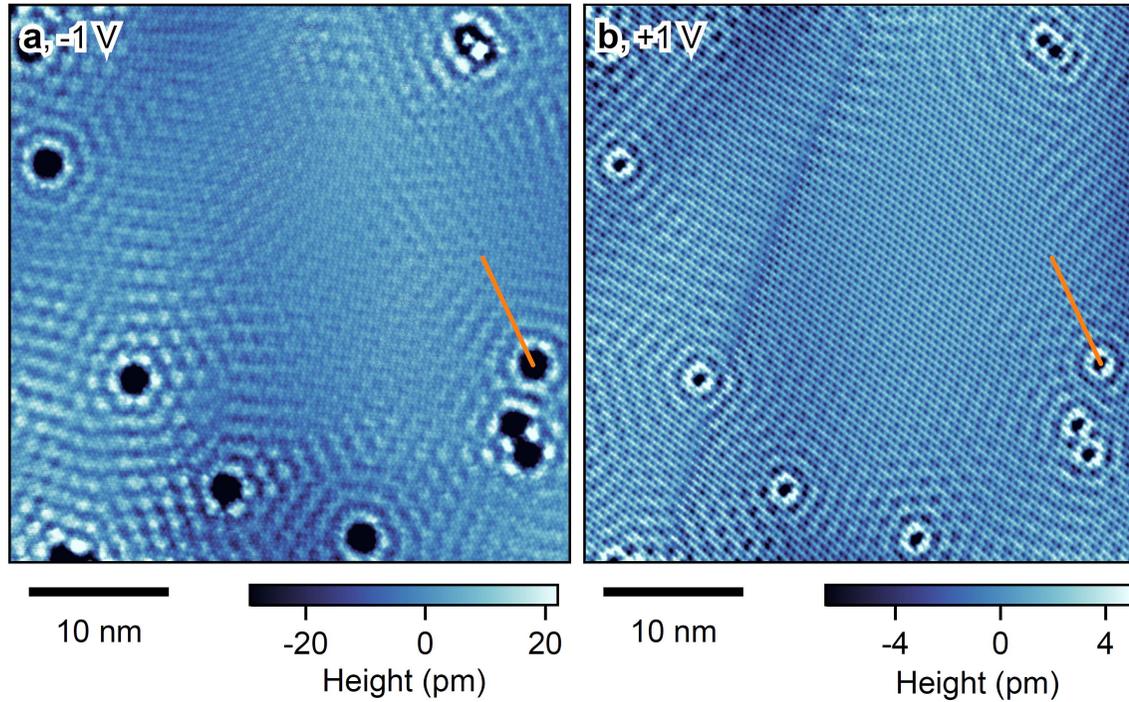
Highly oriented pyrolytic graphite (HOPG)



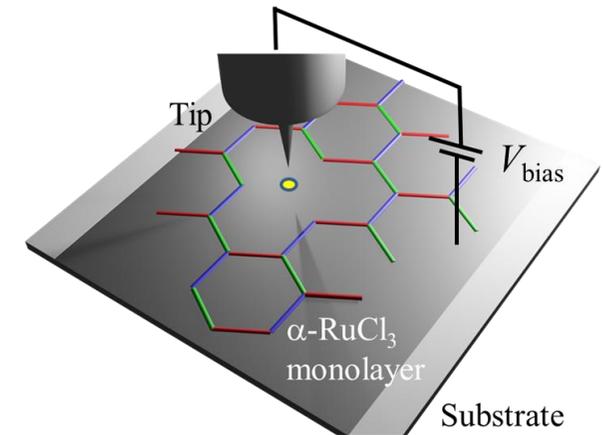
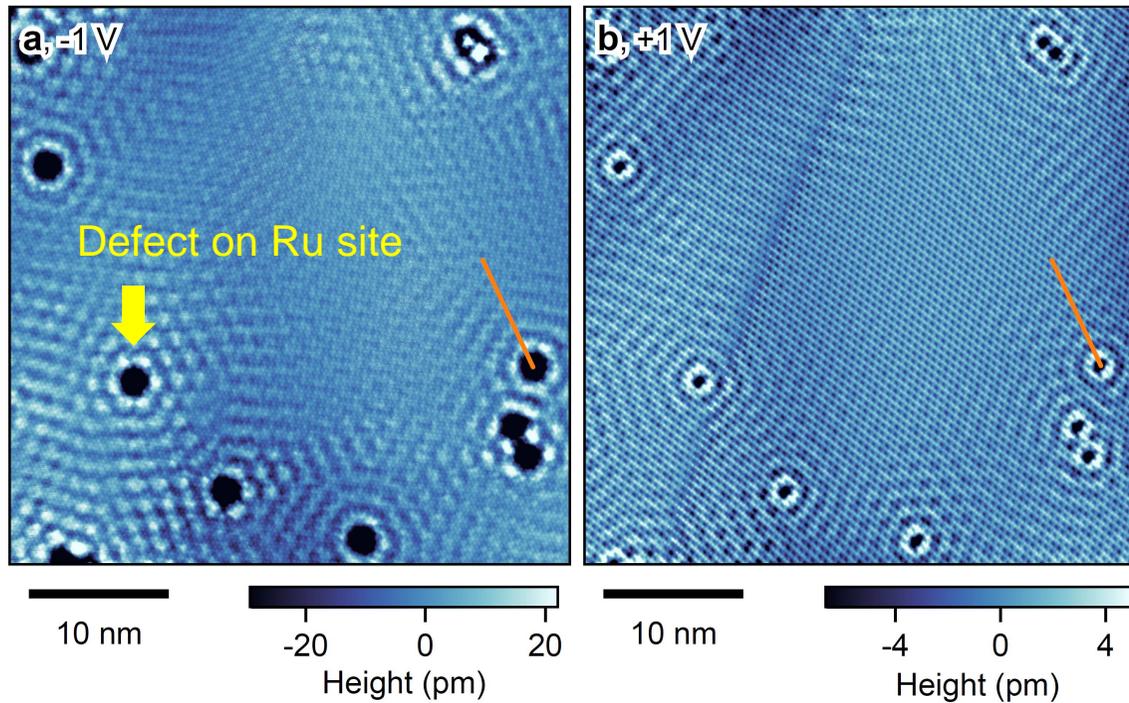
Topographic image (5 K)



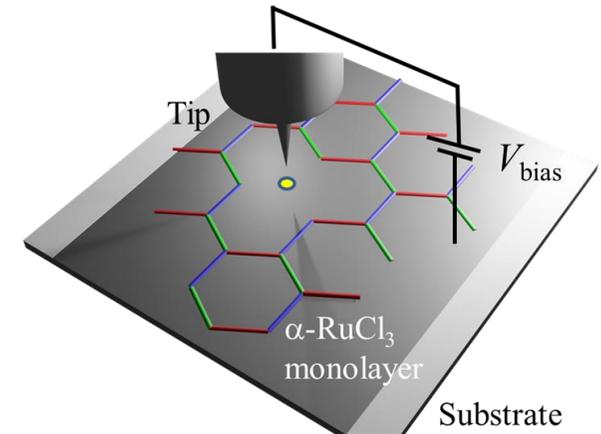
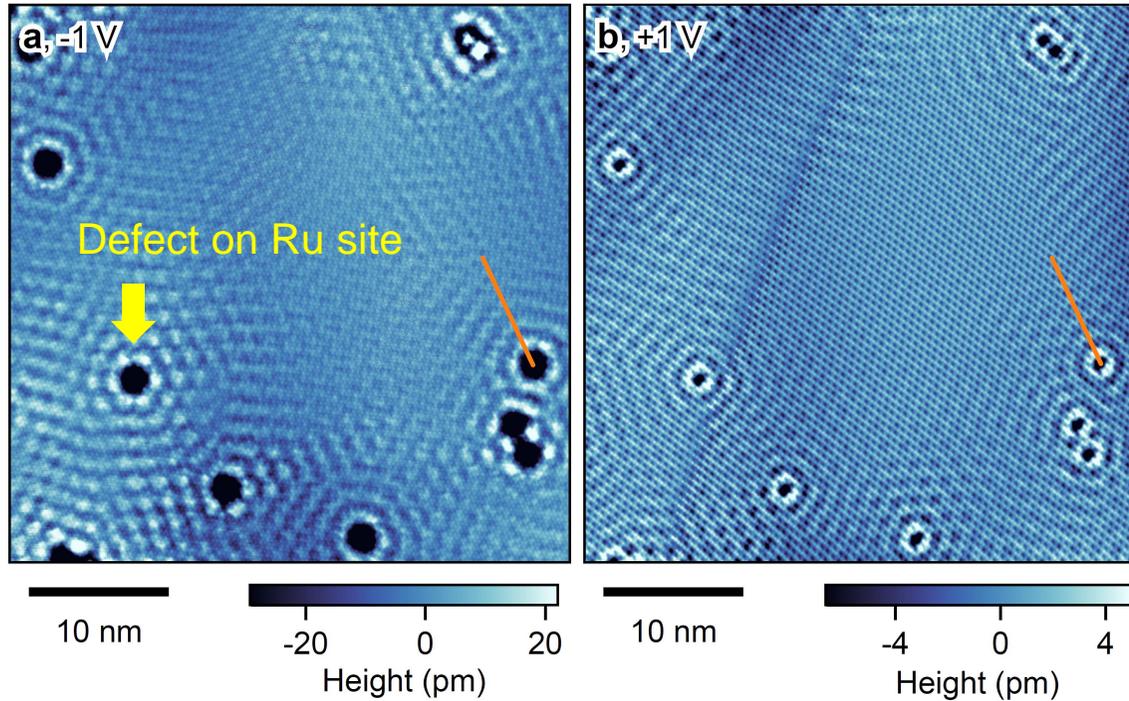
Anomalous spatial oscillations in Kitaev QSL



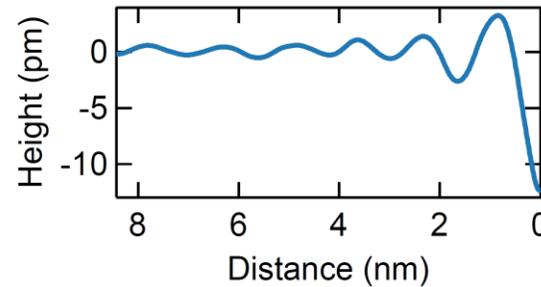
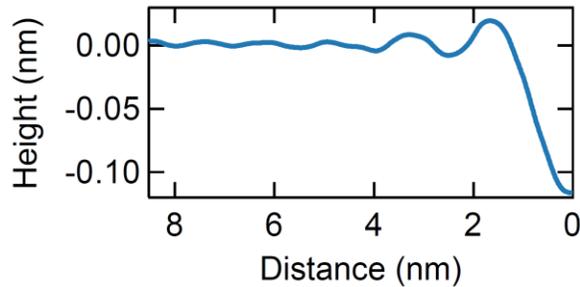
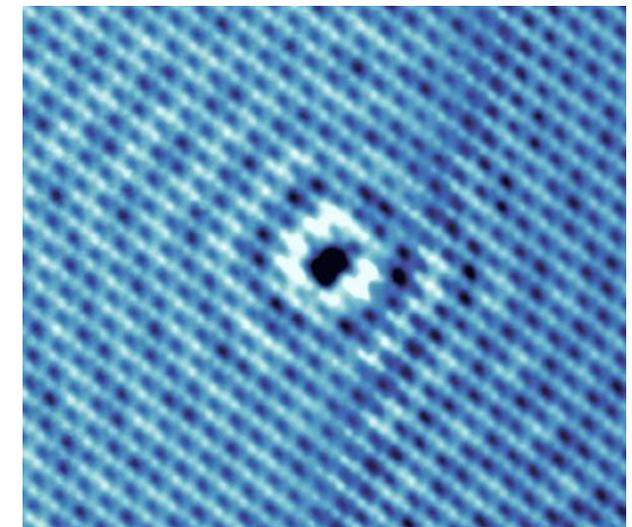
Anomalous spatial oscillations in Kitaev QSL



Anomalous spatial oscillations in Kitaev QSL

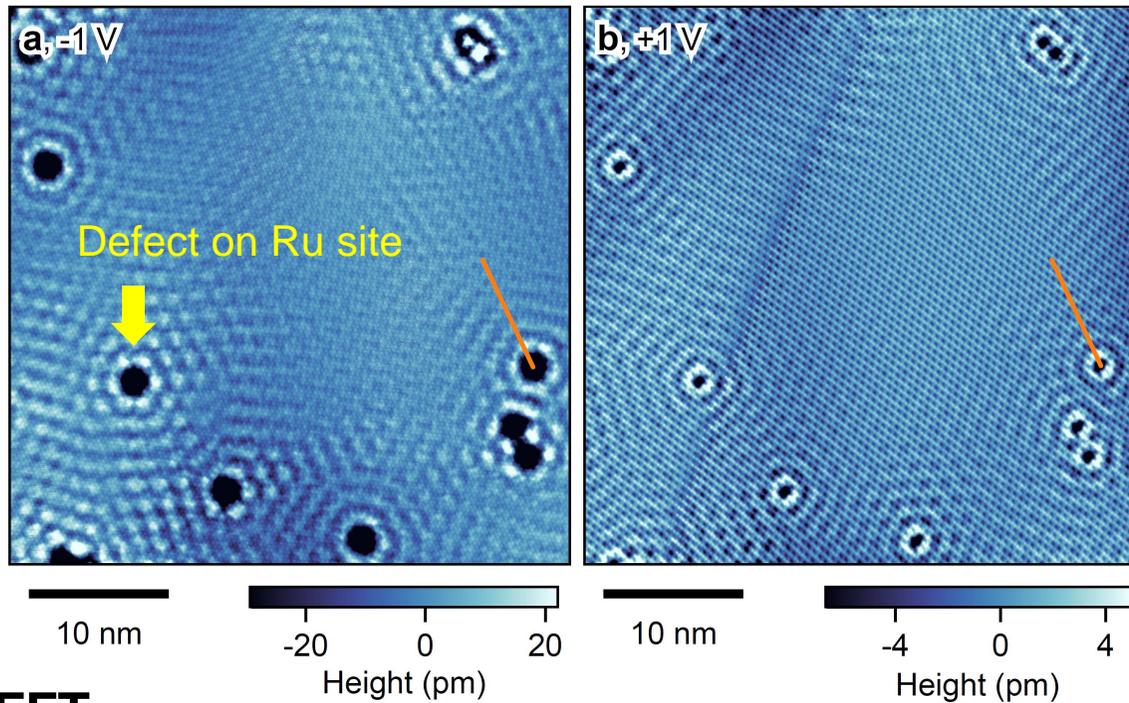


Ripples around defects

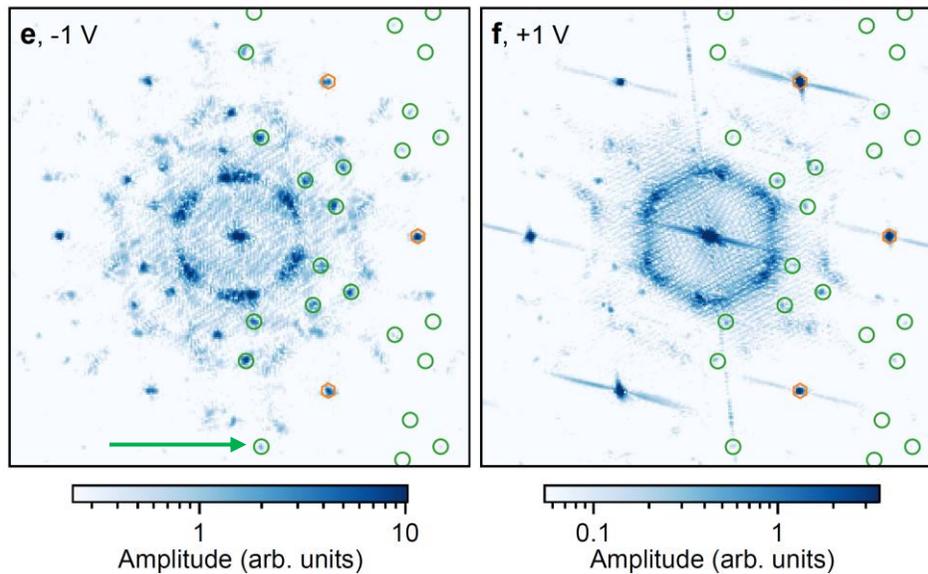


Long-wavelength oscillations around defects
Incommensurate to the lattice spacing

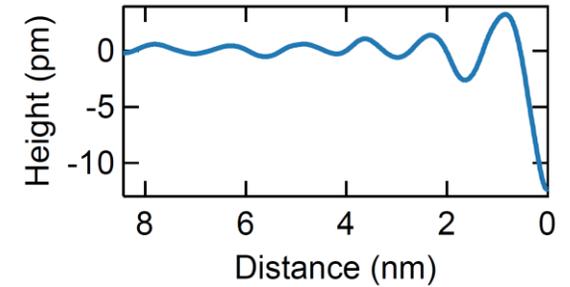
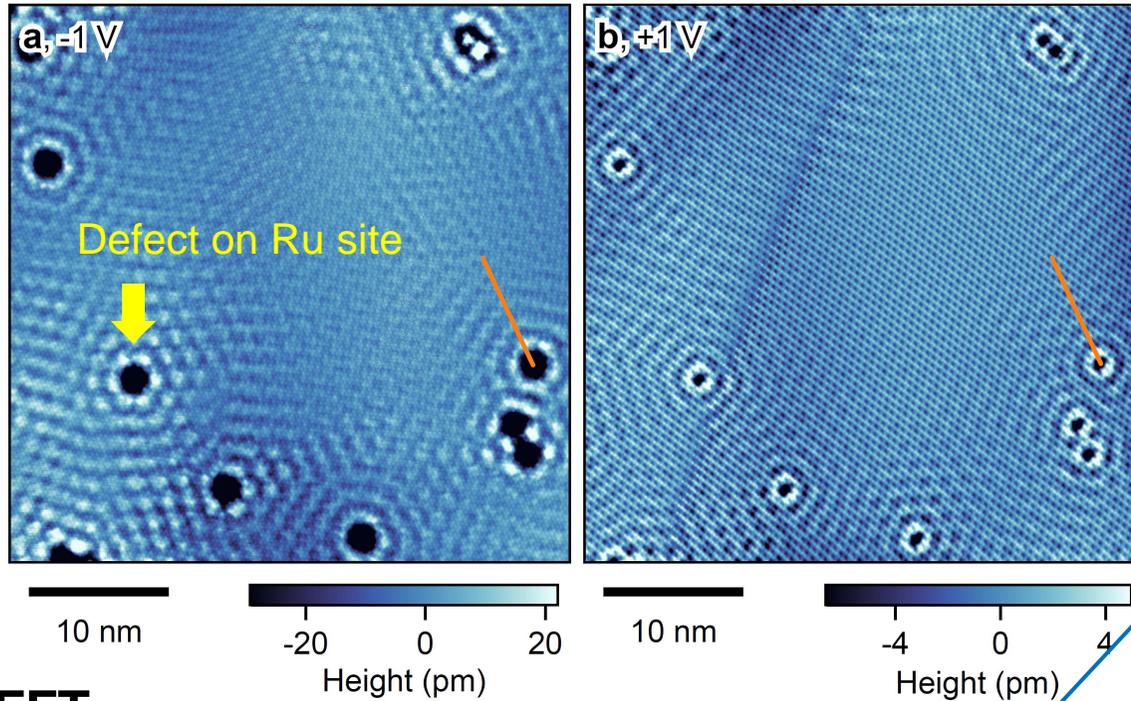
Anomalous spatial oscillations in Kitaev QSL



FFT

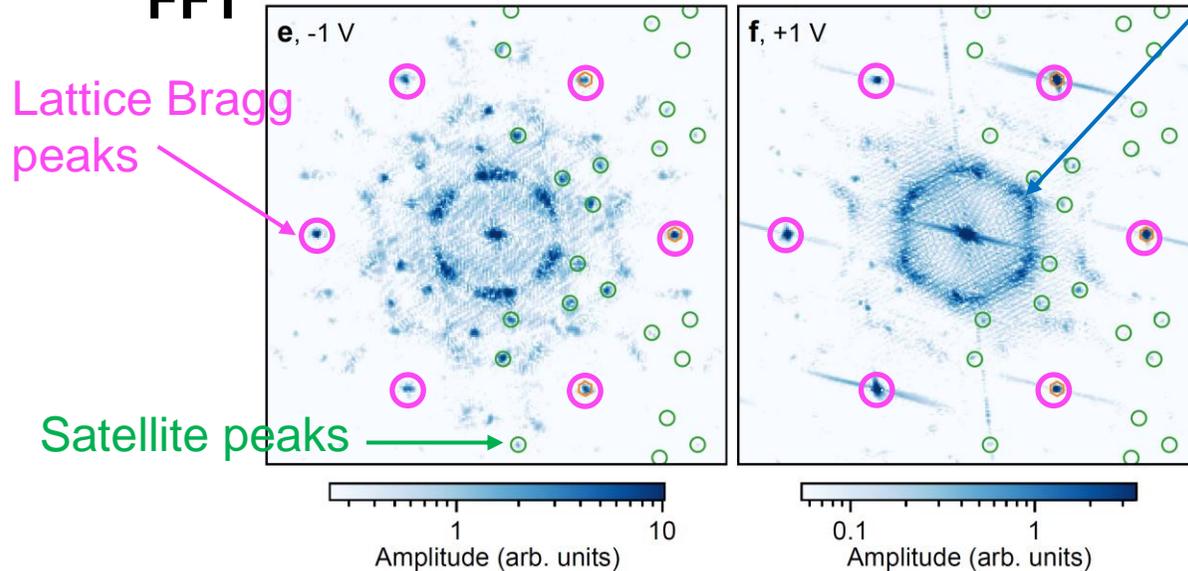


Anomalous spatial oscillations in Kitaev QSL

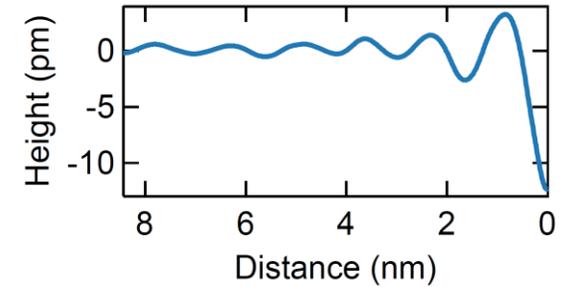
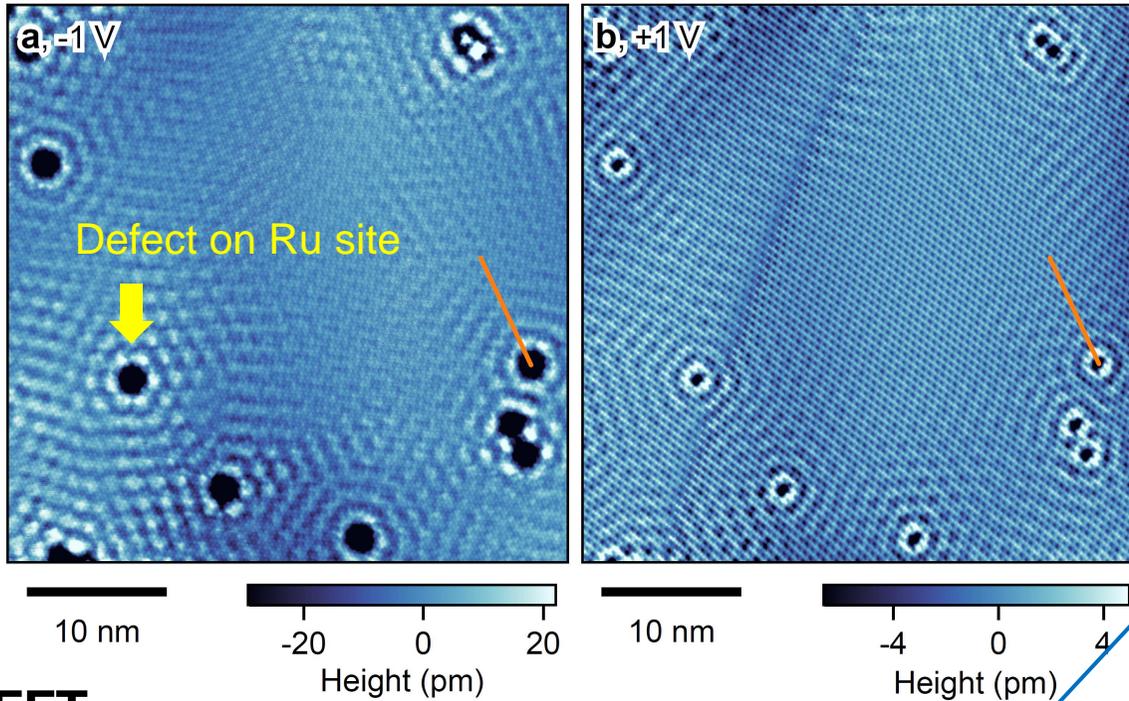


Peaks corresponding to anomalous oscillations around defects

FFT

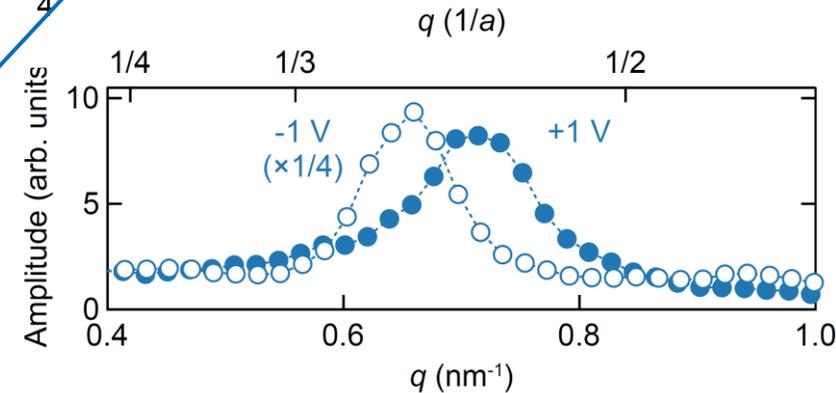
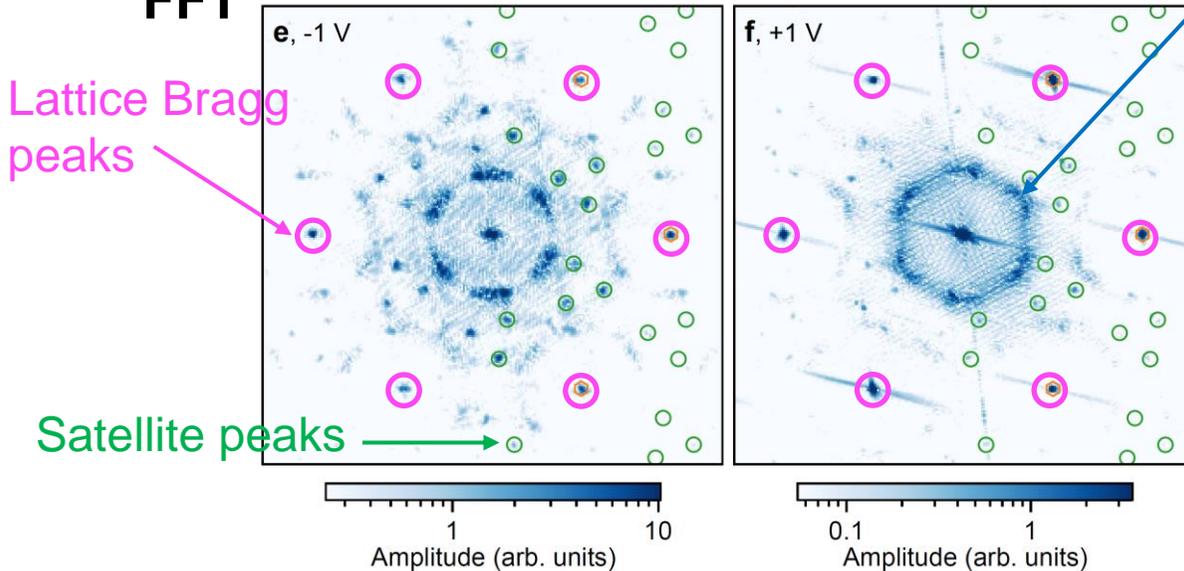


Anomalous spatial oscillations in Kitaev QSL



Peaks corresponding to anomalous oscillations around defects

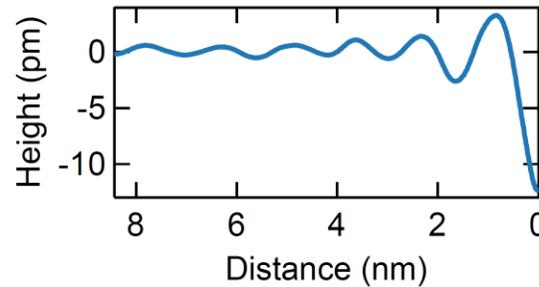
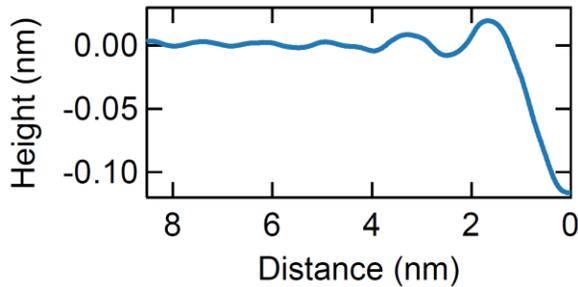
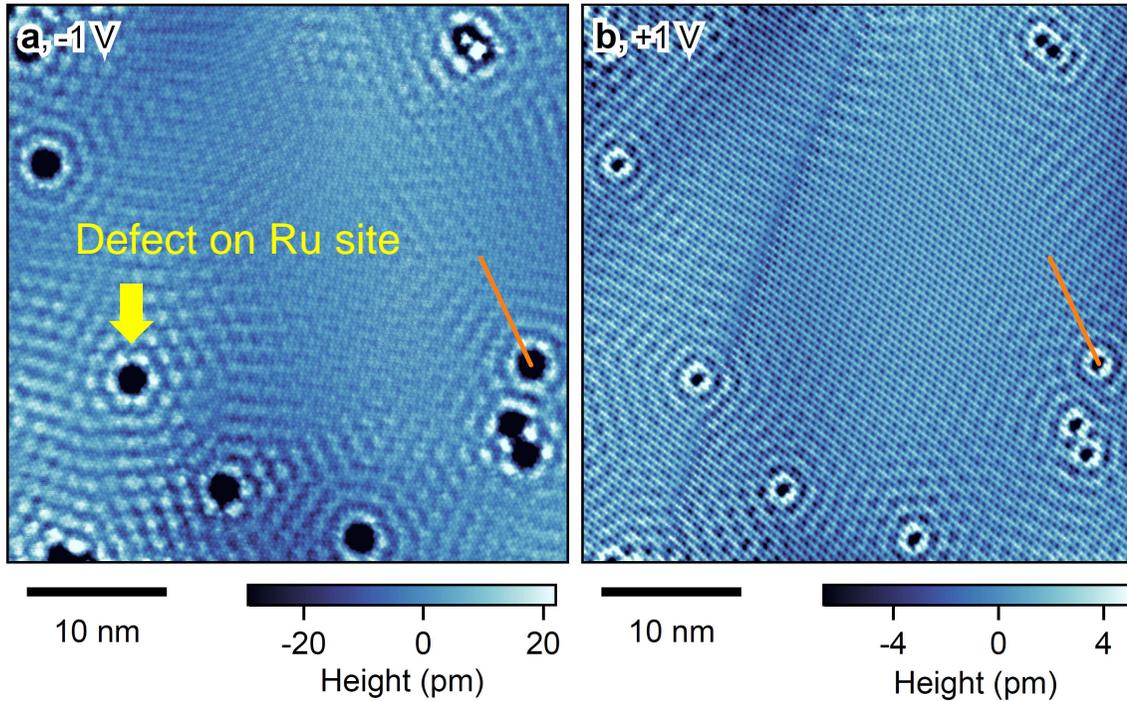
FFT



Period of the oscillations depends on the sign of the bias voltage

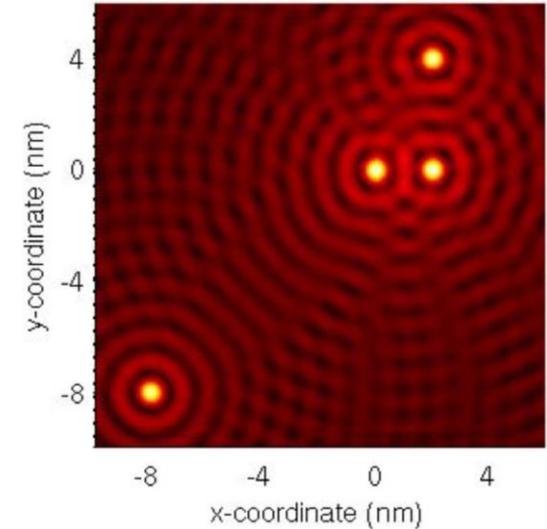
Not CDW or lattice distortion

Anomalous spatial oscillations in Kitaev QSL



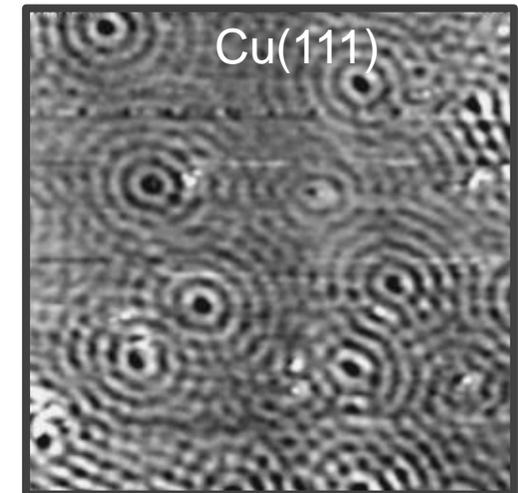
Long-wavelength oscillation around Ru defects
Incommensurate to the lattice spacing

Friedel oscillations (metal)



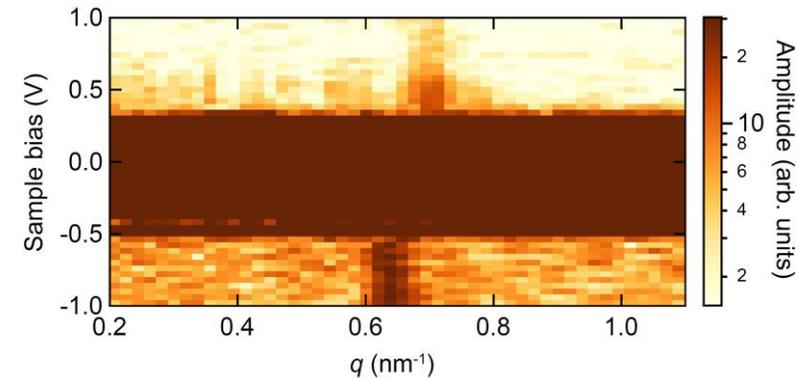
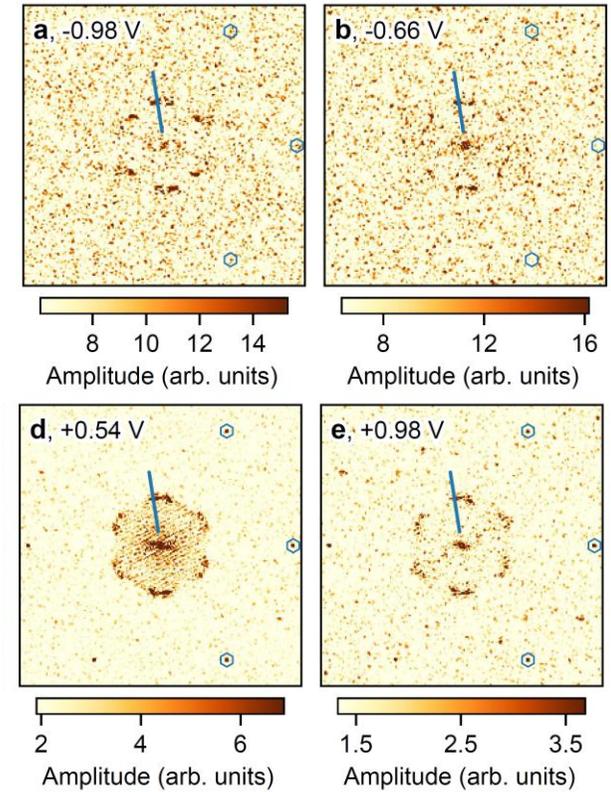
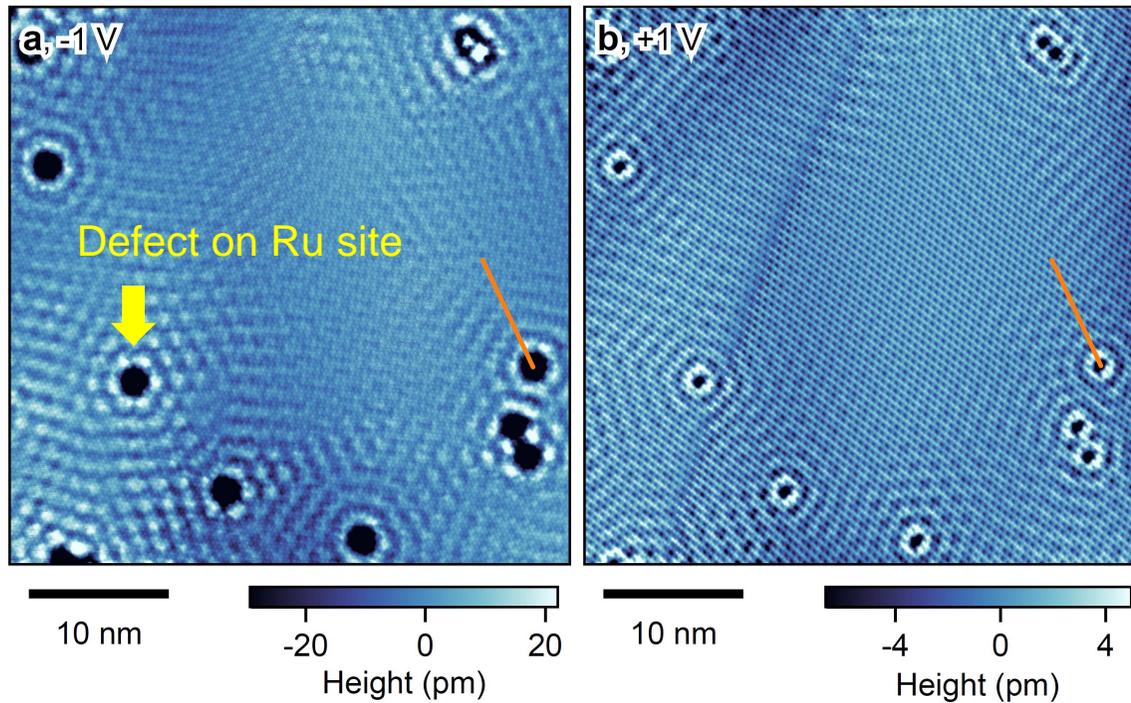
J. Fransson and A. V. Balatsky PRB (2007)

Quasiparticle interference (QPI)



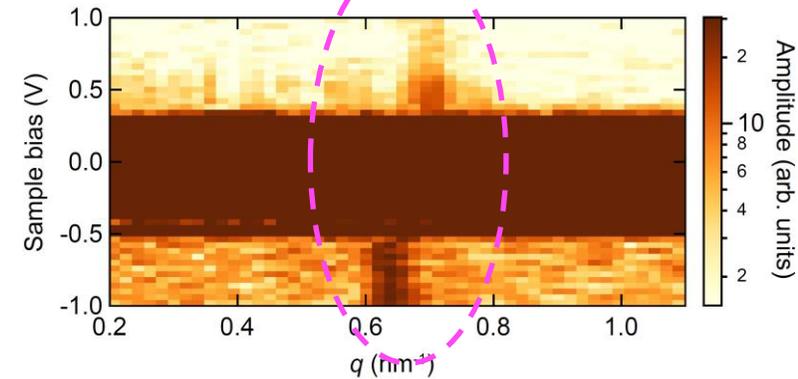
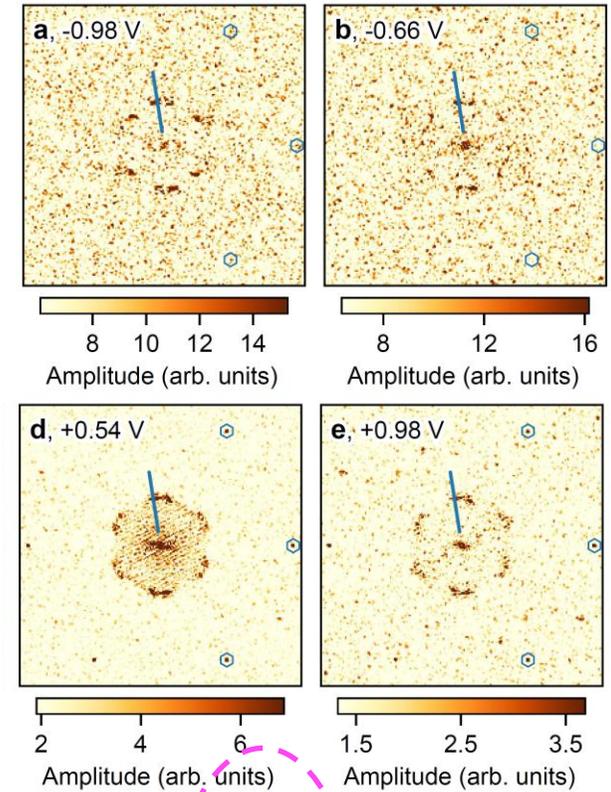
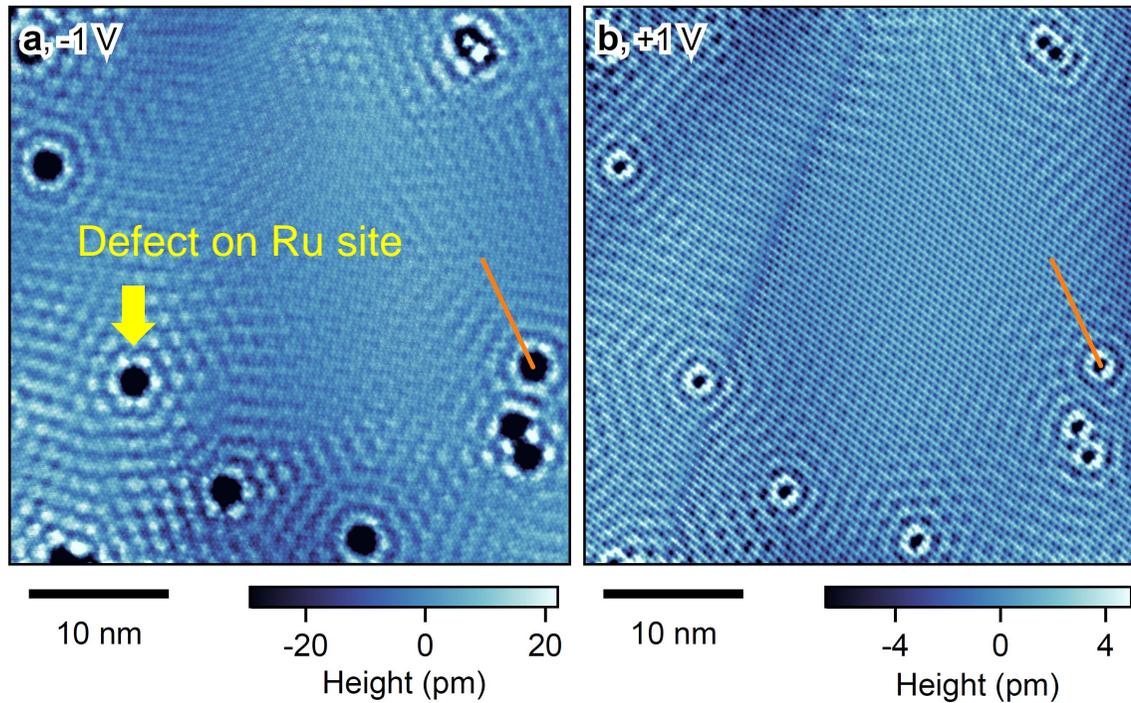
L. Petersen *et al.*, PRB (1998).

Anomalous spatial oscillations in Kitaev QSL

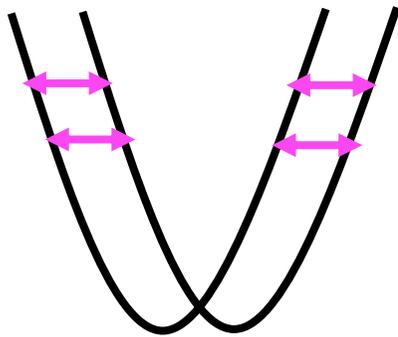


Negligibly small q -dependence
Not QPI

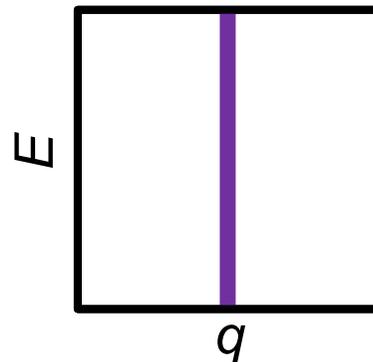
Anomalous spatial oscillations in Kitaev QSL



Destructive interference



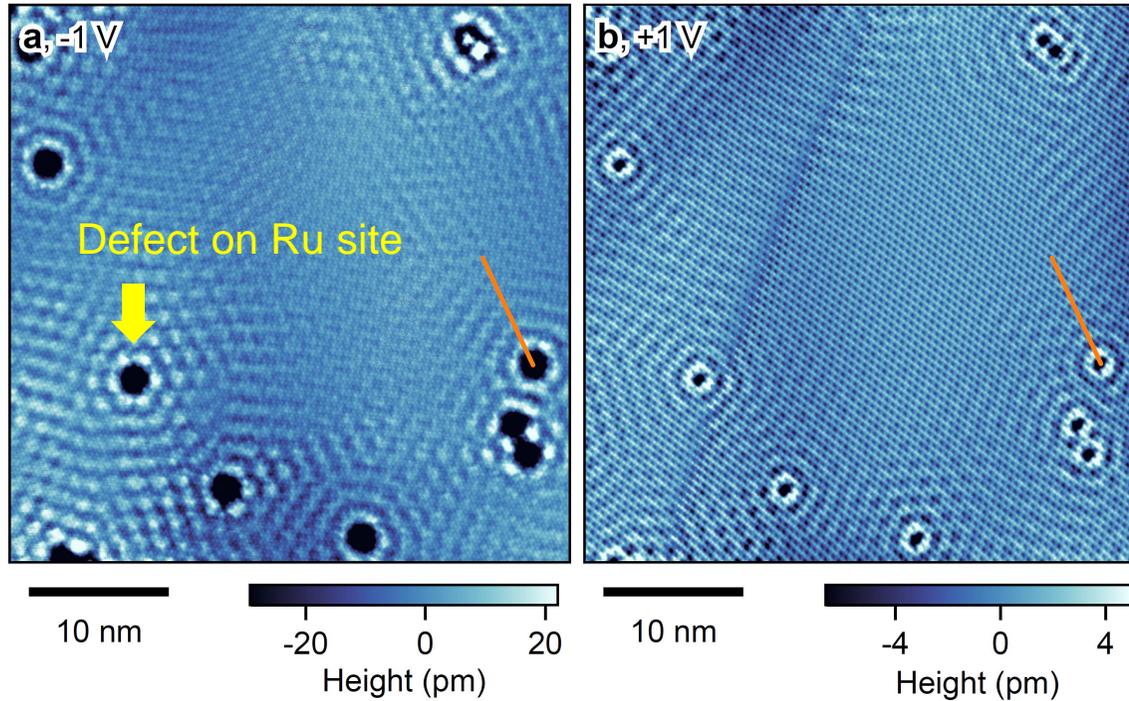
Vertical dispersion does not appear in QPI spectra



Negligibly small q -dependence

Not QPI

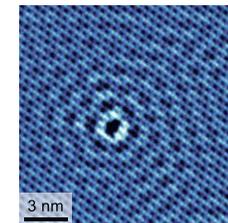
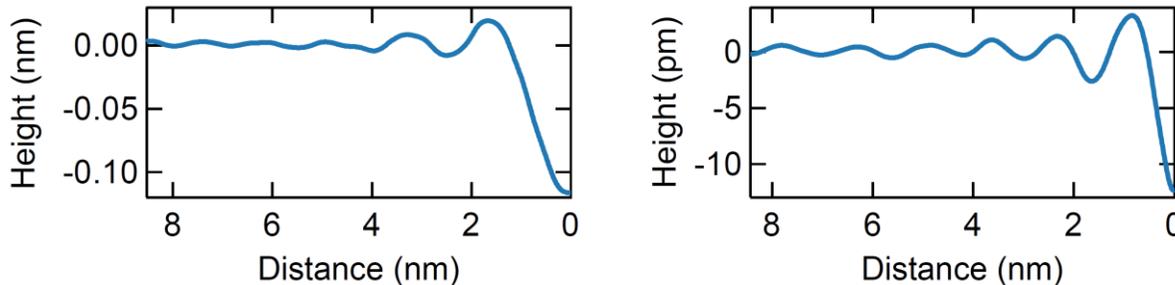
Enigmatic spatial oscillations in Kitaev QSL



Lattice distortion
Moiré
Charge density wave
Friedel oscillations
Quasiparticle Interference

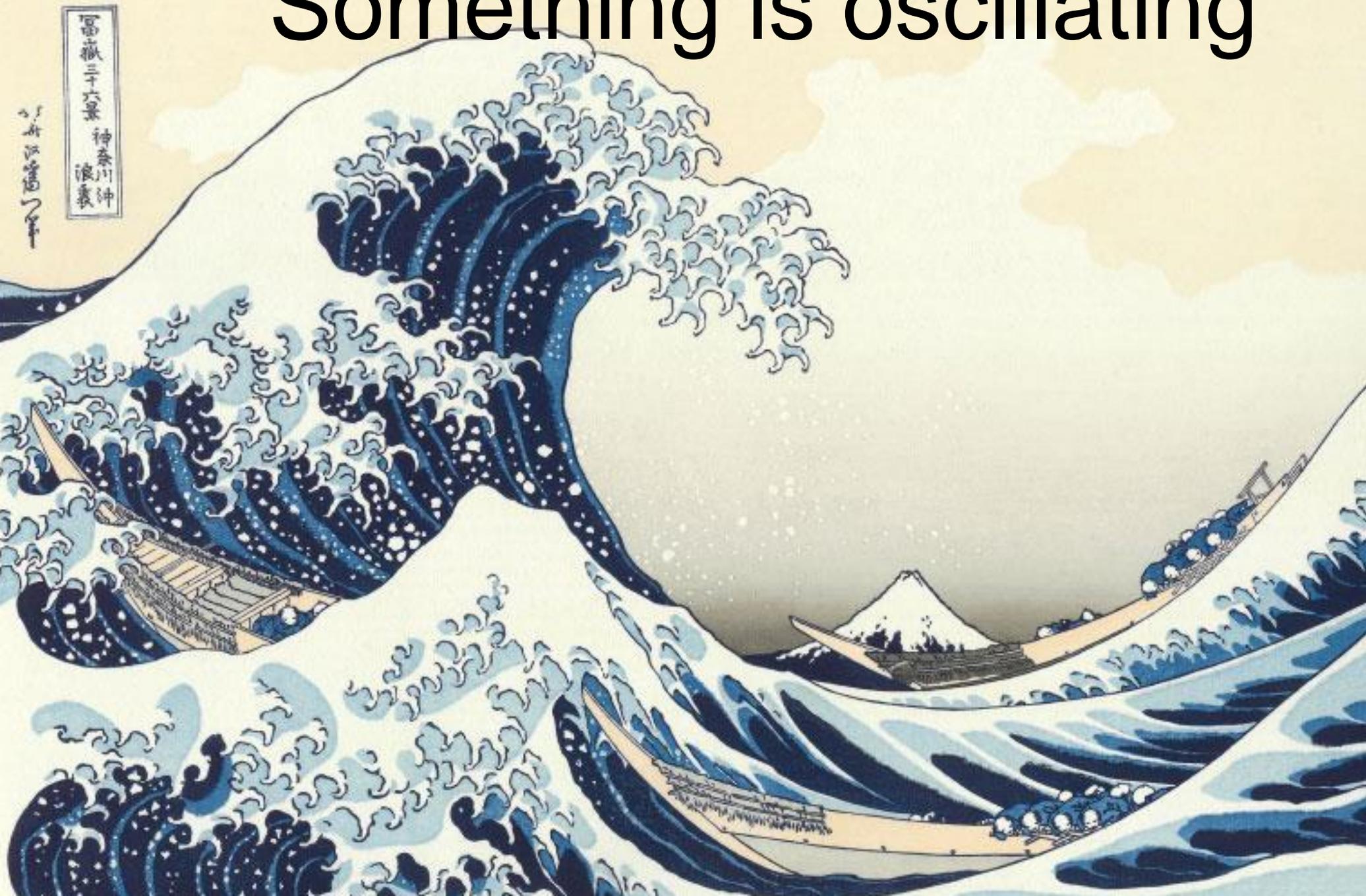
are NOT the origin of the observed spatial oscillations

Interference phenomena related to Majorana excitations?



**Long-wavelength oscillation around Ru defects
Incommensurate to the lattice spacing**

Something is oscillating



Summary

Topology and thermodynamics in α -RuCl₃

- Half-integer quantized thermal Hall effect in α -RuCl₃
 - Y. Kasahara *et al.* Phys. Rev. Lett. **120**, 217205 (2018).
 - Y. Kasahara *et al.* Nature **559**, 227 (2018).
 - Y. Kasahara *et al.* Phys. Rev. B **106**, L060410 (2022).
- Hall sign consistent with topological invariant of KQSL
- Dirac cone for $\mathbf{H} \parallel b$ and gap Δ_M for $\mathbf{H} \parallel a$ ($\Delta_M \propto H^3$)
 - T. Yokoi *et al.* Science **373**, 568 (2021).
 - O. Tanaka *et al.* Nature Phys. **18**, 429 (2022).
- Sign change of the planar thermal Hall effect and gap closing occur simultaneously at $\mathbf{H} \parallel b$
 - K. Imamura, S. Suetsugu *et al.* arXiv:2305.10619.

Fermion, not boson

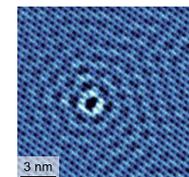
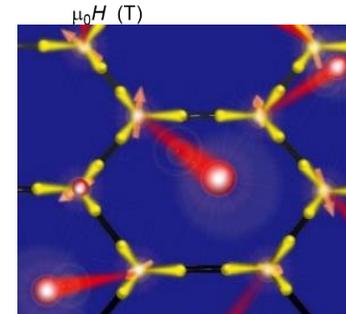
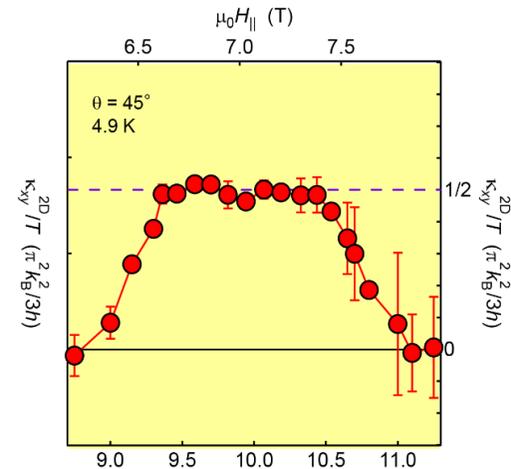
No κ_{xx} oscillations in very clean crystals

Majorana fermion is the origin of thermal Hall effect

STM studies on α -RuCl₃ monolayer

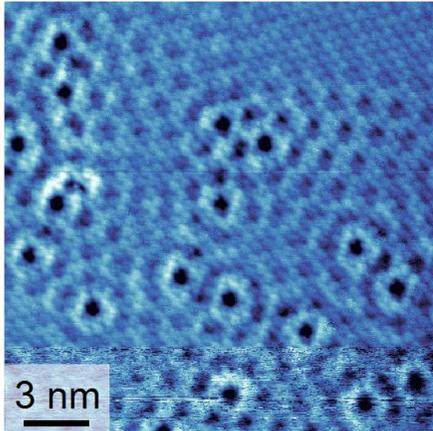
Enigmatic spatial oscillations around defects

Y. Kohsaka *et al.* a preprint.

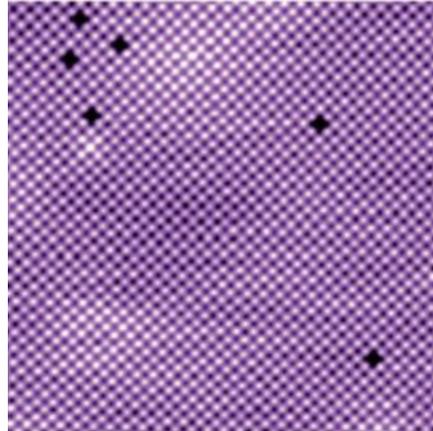


Periodic long-wavelength structure

α -RuCl₃

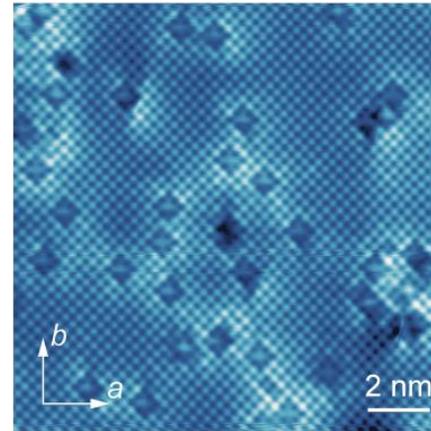


Ca₂CuO₂Cl₂



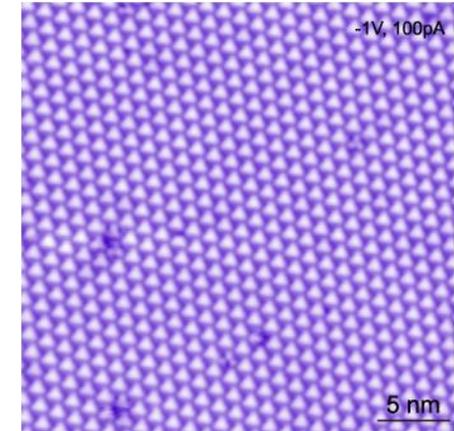
C. Ye *et al.*, *Nat. Commun.*
4, 1365 (2013).

Sr₂IrO₄



Z. Sun *et al.*, *Phys. Rev. Res.*
3, 023075 (2021).

1T-TaS₂

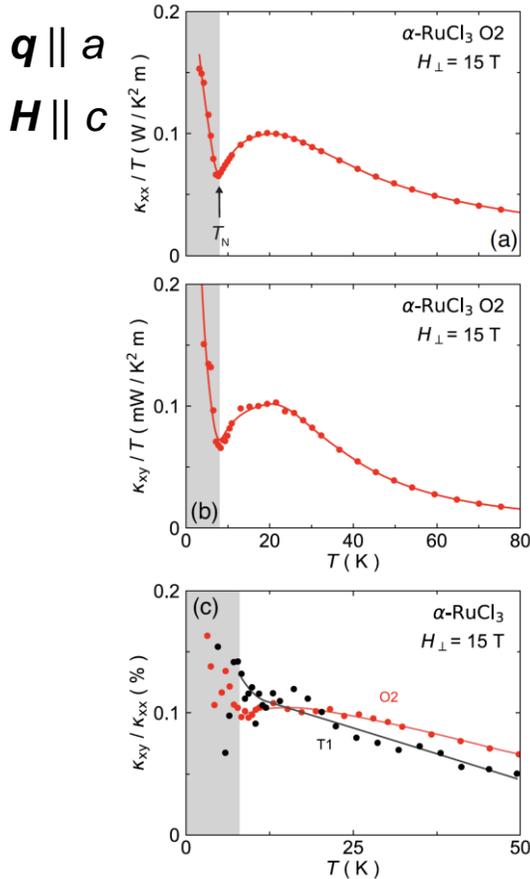


Z. Wu *et al.*, *Phys. Rev. B*
105, 035109 (2022).

No long-wavelength oscillations have been reported in other Mott insulators

Evidence against phonon thermal Hall effect

Sherbrooke



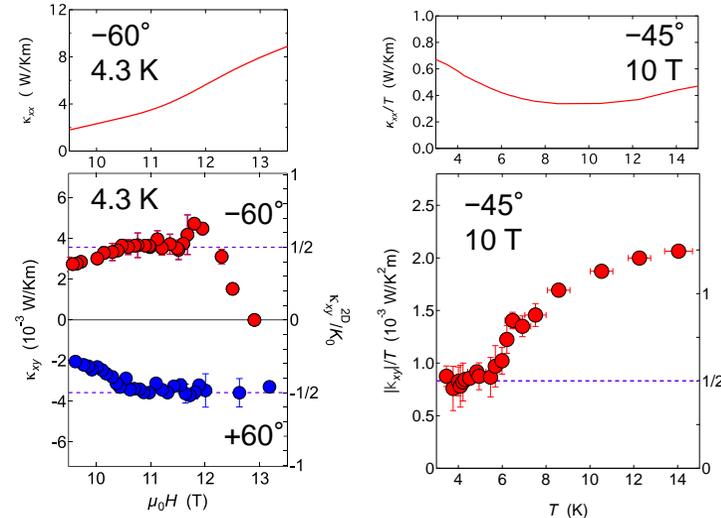
Scaling between κ_{xx} and κ_{xy}

Phonon origin of κ_{xy} ?

É. Lefrançois *et al.*, PRX **12**, 021025 (2022).

Kyoto

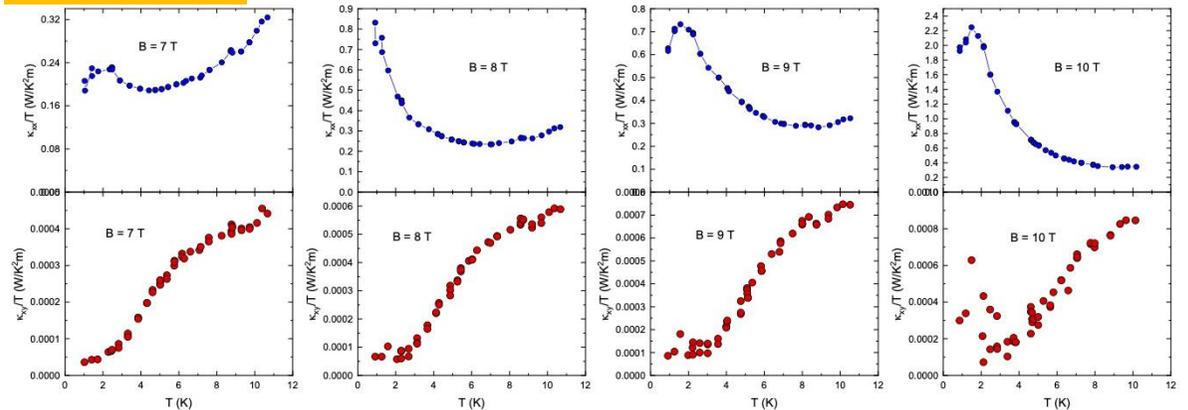
$q \parallel a$



T. Yokoi *et al.*, Science **373**, 568 (2021).

Princeton

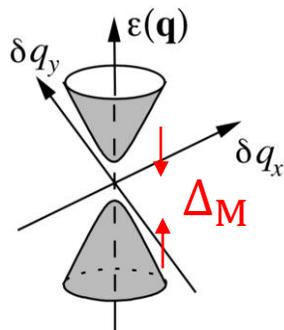
$q \parallel H \parallel a$



P. Czajka *et al.*, Nat. Mater. **22**, 36 (2023).

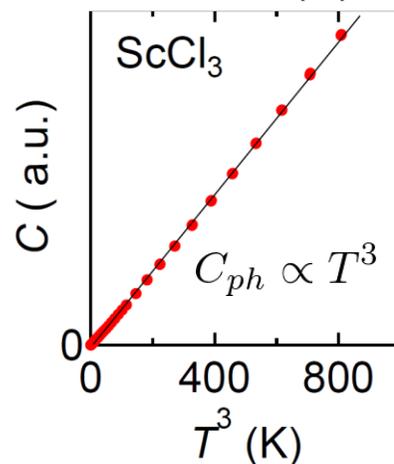
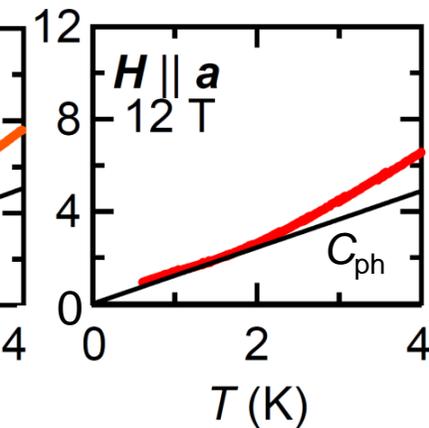
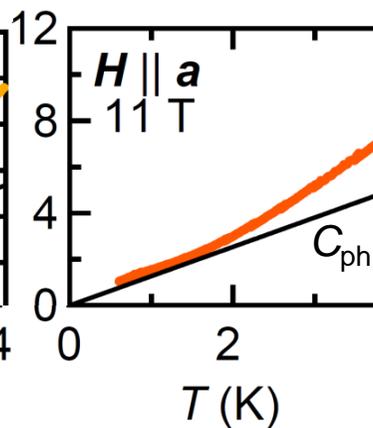
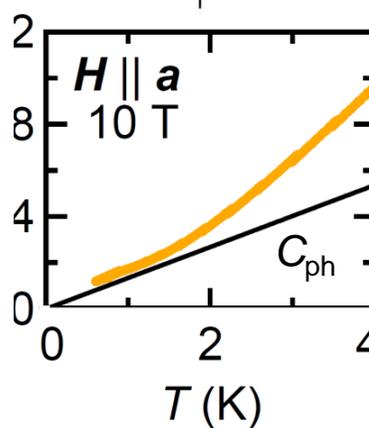
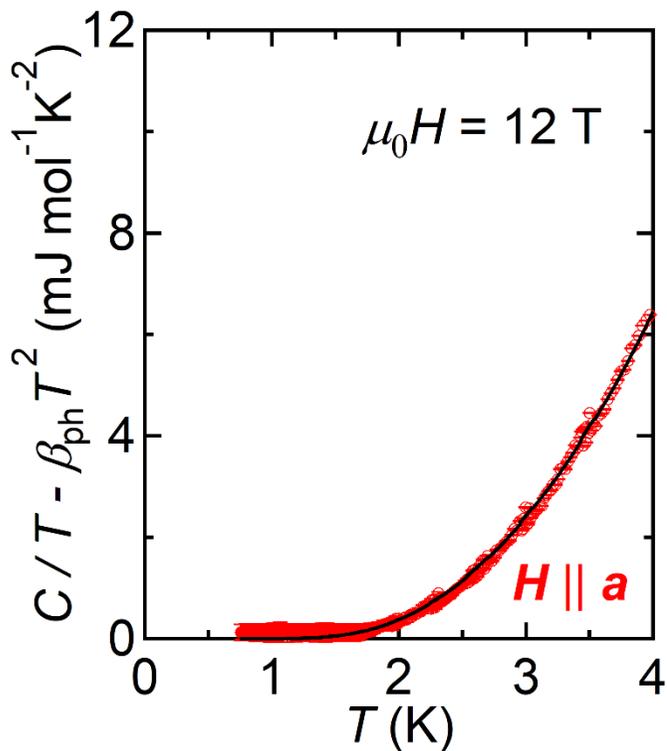
κ_{xx} and κ_{xy} show essentially different T - & H -dependencies.

Majorana gap for $H//a$: specific heat in FIQD state



$$C_{\text{mag}}/T \propto e^{-\frac{\Delta_M}{T}}$$

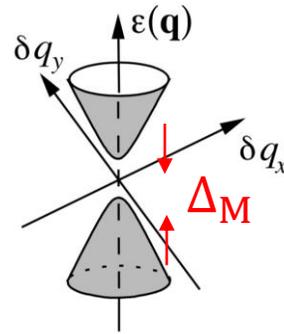
Bulk gap



Nonmagnetic reference

O. Tanaka *et al.* Nature Phys. **18**, 429 (2022).

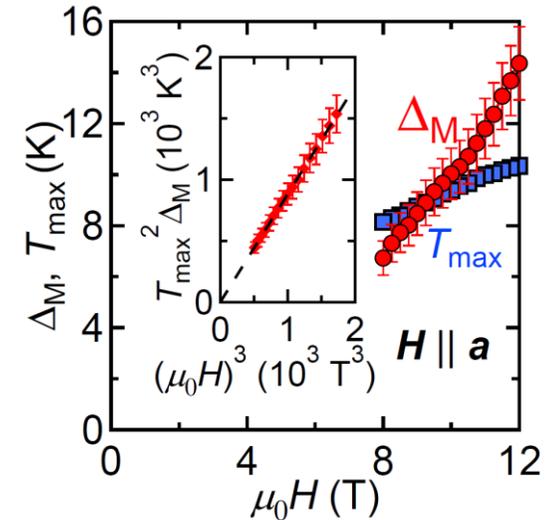
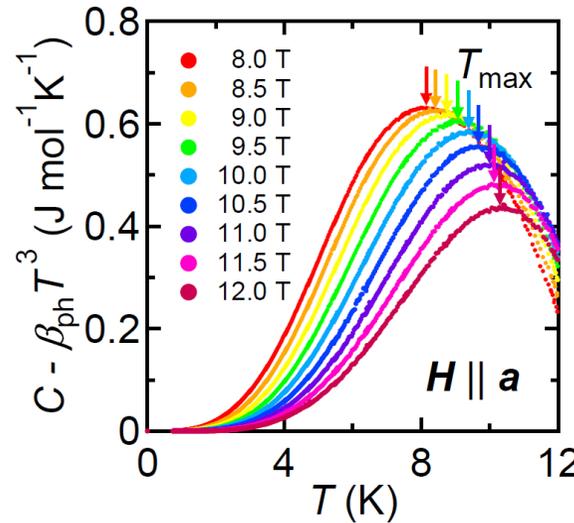
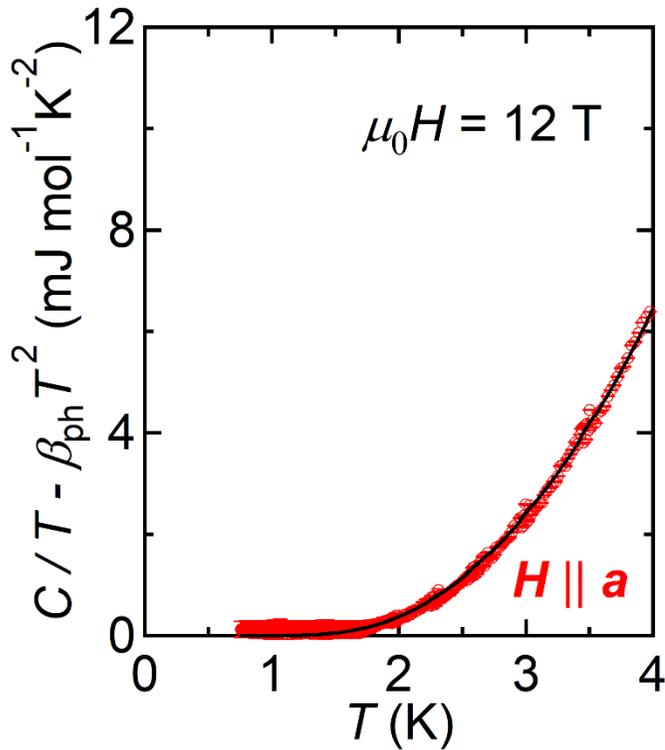
Majorana gap for $H//a$: specific heat in FIQD state



$$C_{\text{mag}}/T \propto e^{-\frac{\Delta_M}{T}}$$

Bulk gap

Δ_M : Majorana gap



$$T_{\text{max}} \propto \Delta_{\text{flux}}$$

Flux (vison) gap

$$\Delta_M T_{\text{max}}^2 \propto H^3$$

Y. Motome and J. Nasu, JPSJ (2020).

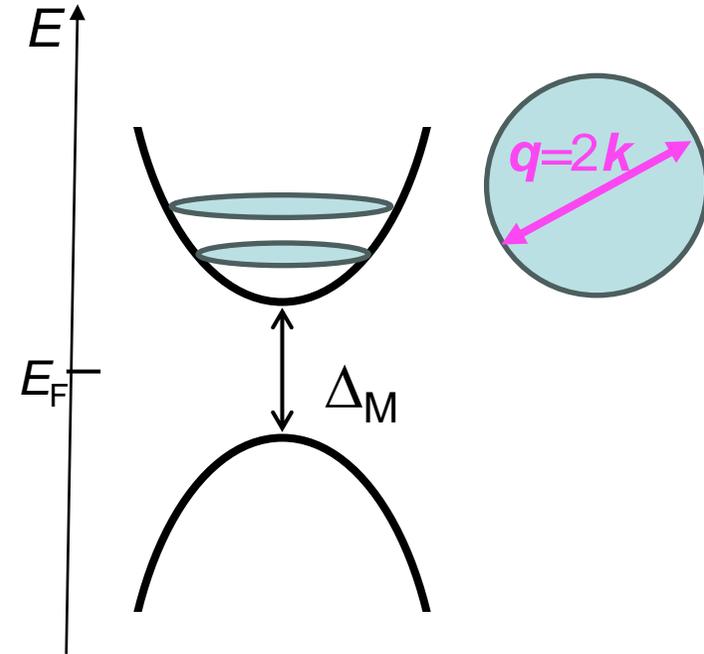
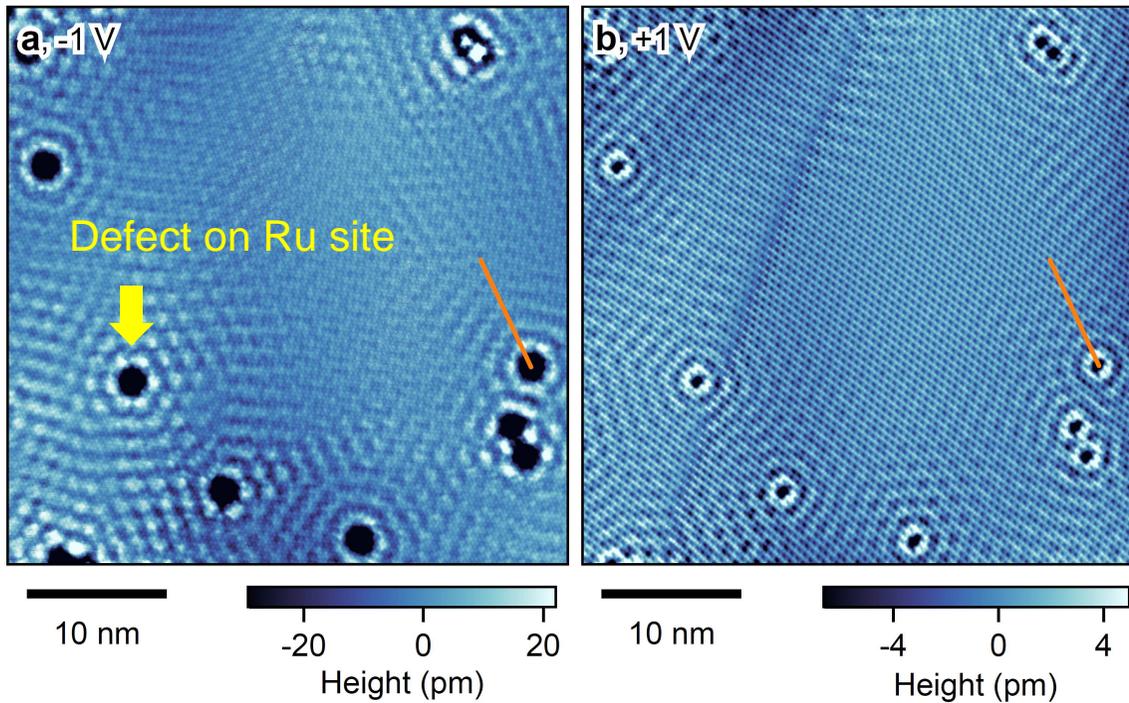
$$\Delta_M \approx \frac{|h_x h_y h_z|}{\Delta_{\text{flux}}^2}$$

3rd order perturbation
opens Majorana gap

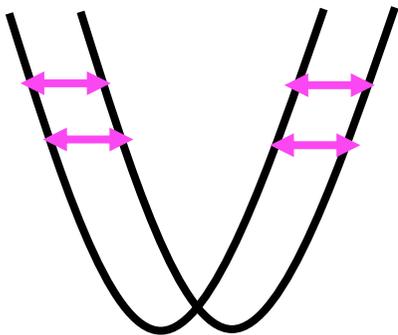
Majorana gap

O. Tanaka *et al.* Nature Phys. **18**, 429 (2022).

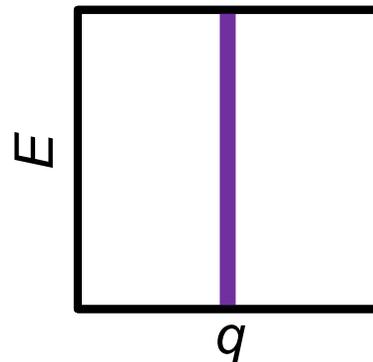
Anomalous spatial oscillations in Kitaev QSL



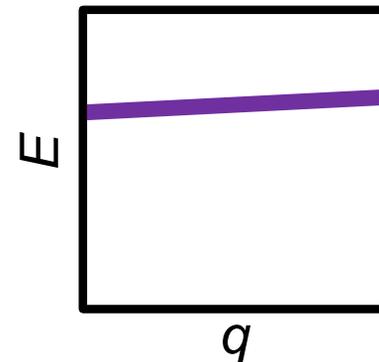
Destructive interference



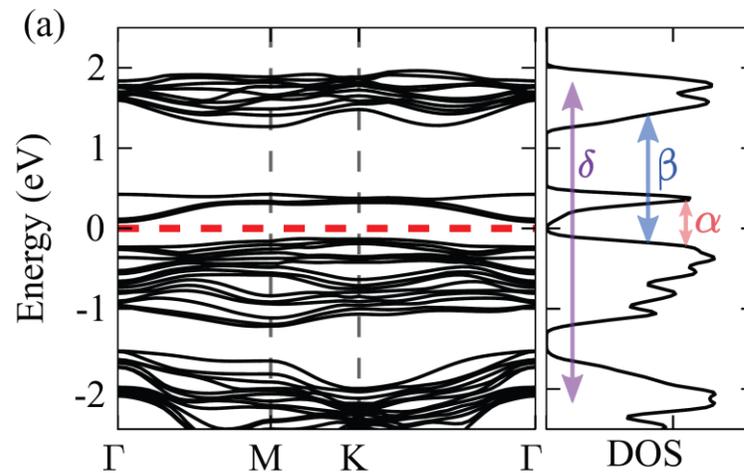
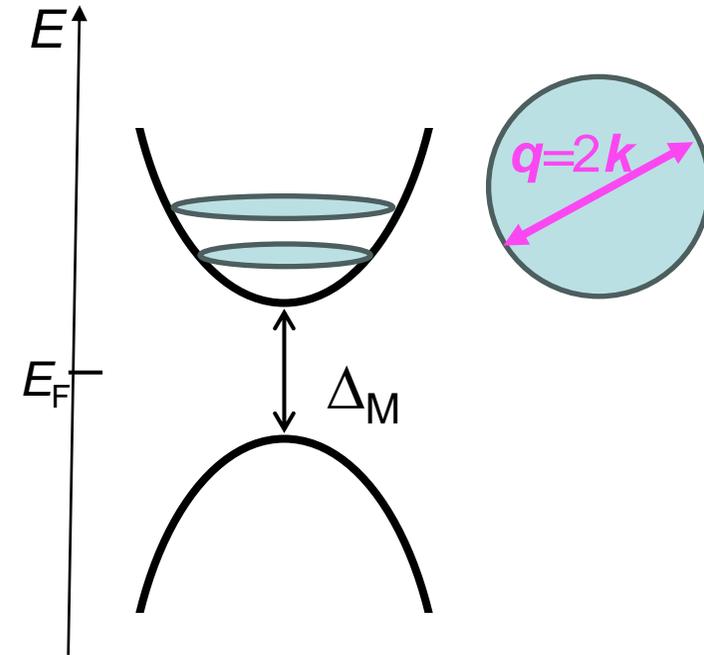
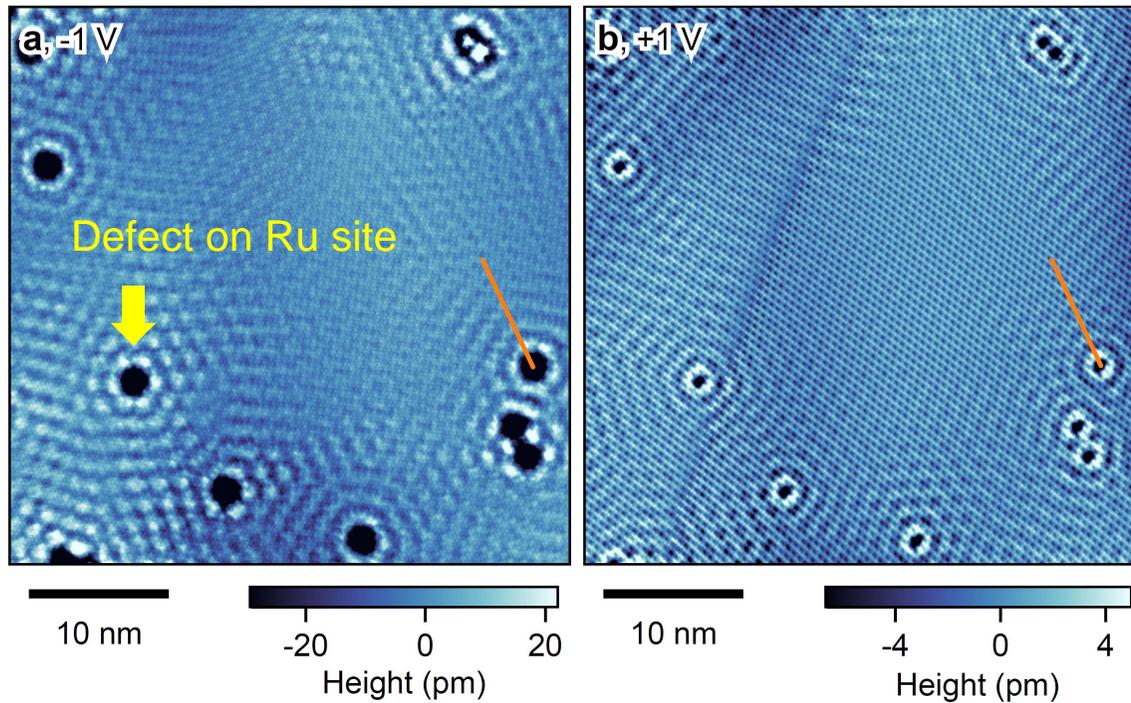
Vertical dispersion does not appear in QPI spectra



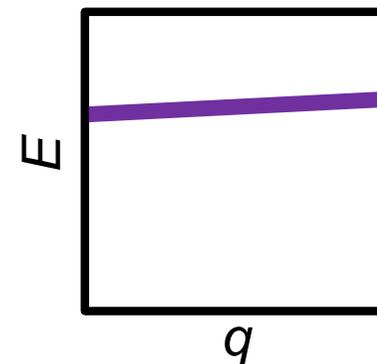
Expected QPI spectra



Anomalous spatial oscillations in Kitaev QSL

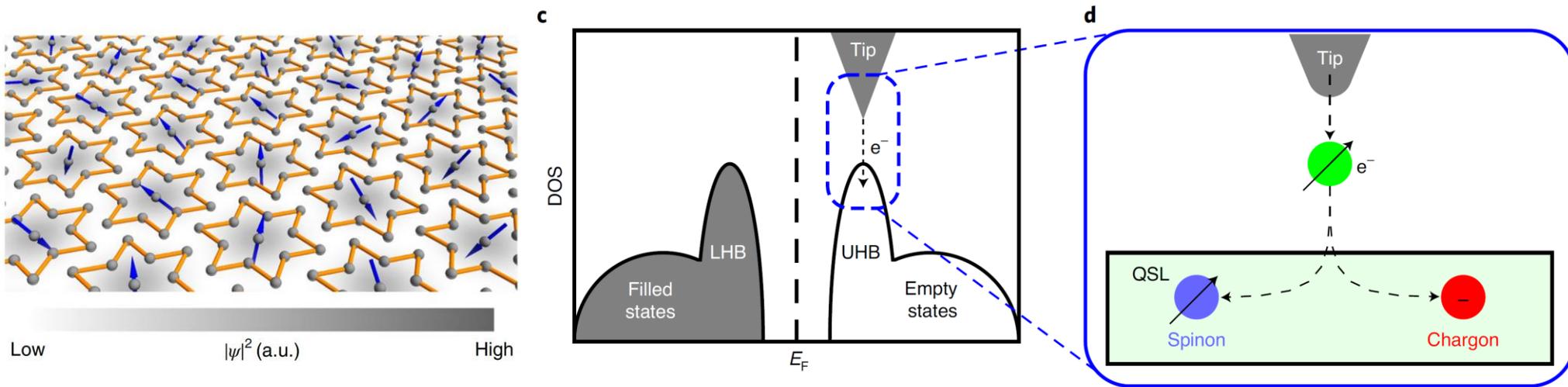


Expected QPI spectra



Evidence for quantum spin liquid behaviour in single-layer 1T-TaSe₂ from scanning tunnelling microscopy

Wei Ruan ^{1,2,3,16}, Yi Chen ^{1,2,16}, Shujie Tang ^{4,5,6,7,8}, Jinwoong Hwang ^{4,6,9}, Hsin-Zon Tsai^{1,10}, Ryan L. Lee², Meng Wu ^{1,2}, Hyejin Ryu^{6,11}, Salman Kahn ², Franklin Liou², Caihong Jia^{1,2,12}, Andrew Aikawa², Choongyu Hwang ⁹, Feng Wang ^{1,2,13}, Yongseong Choi ¹⁴, Steven G. Louie ^{1,2}, Patrick A. Lee¹⁵, Zhi-Xun Shen ^{4,5}, Sung-Kwan Mo ⁶ and Michael F. Crommie ^{1,2,13} ✉

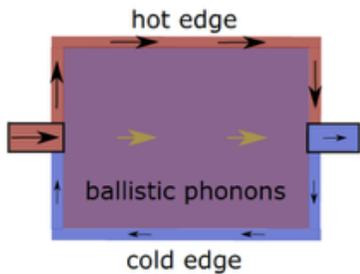
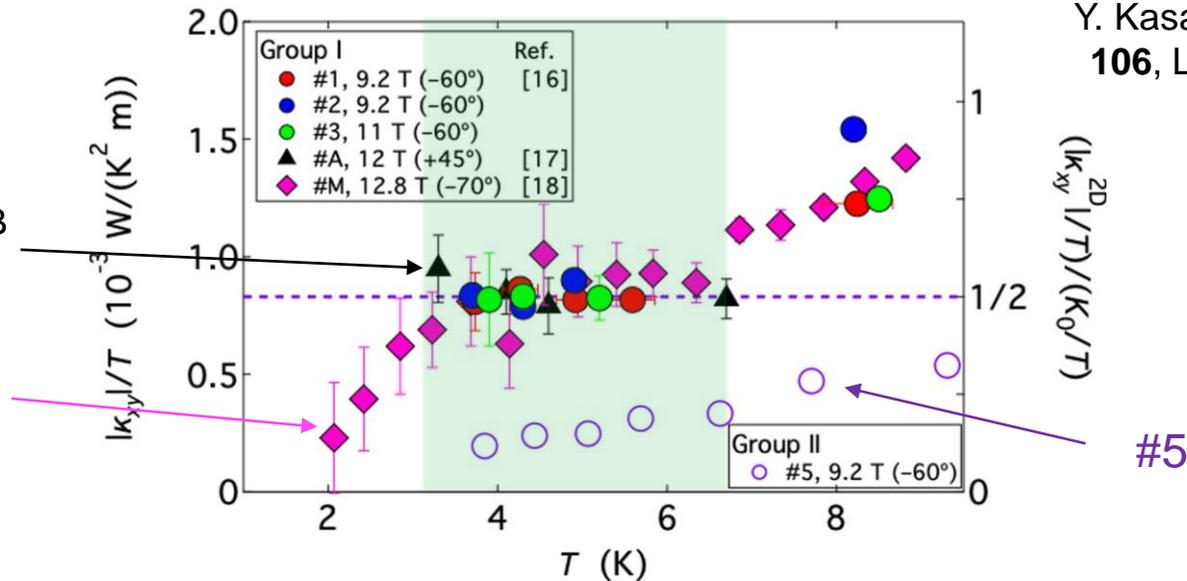


Quantized and unquantized thermal Hall effect

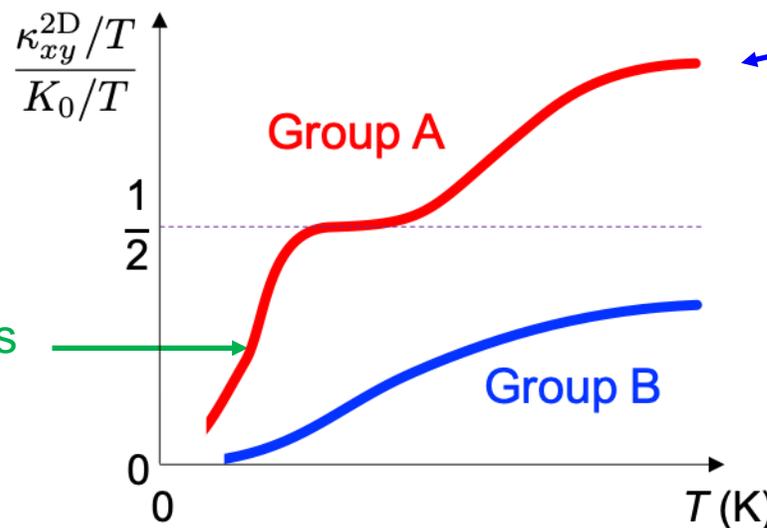
Y. Kasahara, *et al.* PRB **106**, L060410 (2022).

M. Yamashita *et al.* PRB **102**, 220404(R) (2020).

J.A.N. Bruin *et al.* Nature Phys. **18**, 401 (2022).



Decoupling of phonons and edge currents (sample size $\sim \ell_{ph}$)



Majorana+Vison

A.P. Joy and A. Rosch, arXiv:2109.00250

Different types of Majorana excitations

A. Go.J. Jung and E-G. Moon, PRL **122**, 147203 (2019).

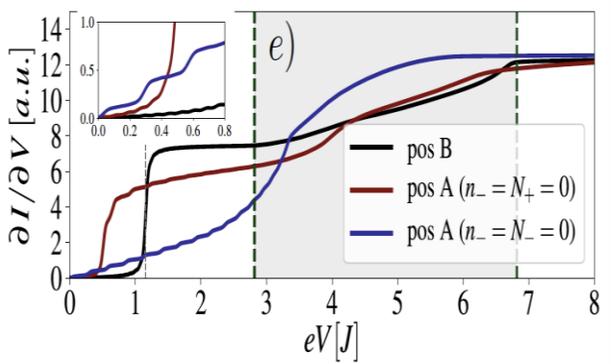
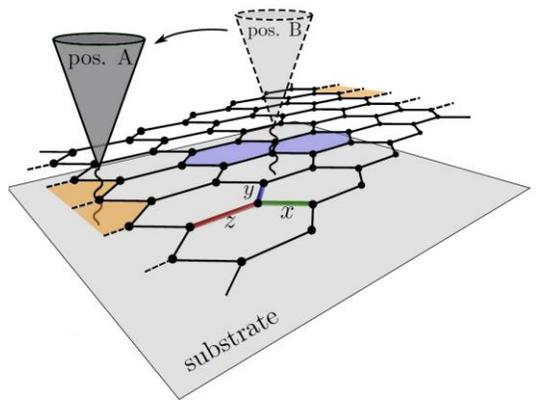
Y. Vinkler-Aviv and A. Rosch, PRX **8**, 021032 (2018).

M.Ye, *et al.*, PRL **121**, 147201 (2018).

Tunneling spectroscopy of Kitaev QSL using monolayer films

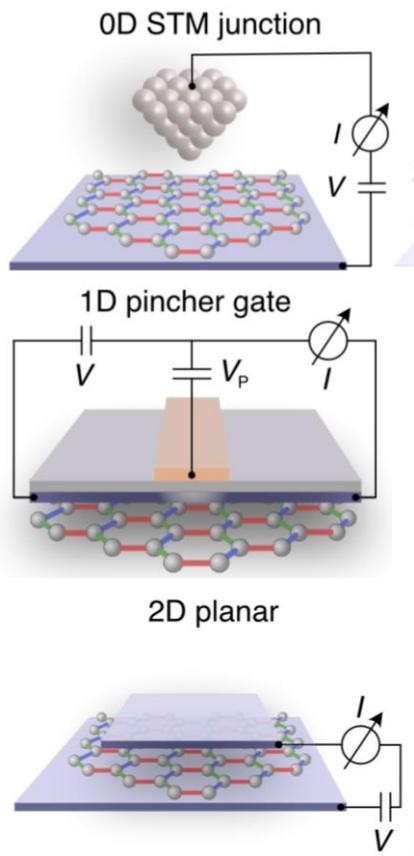
Detections of

Charge-neutral edge state



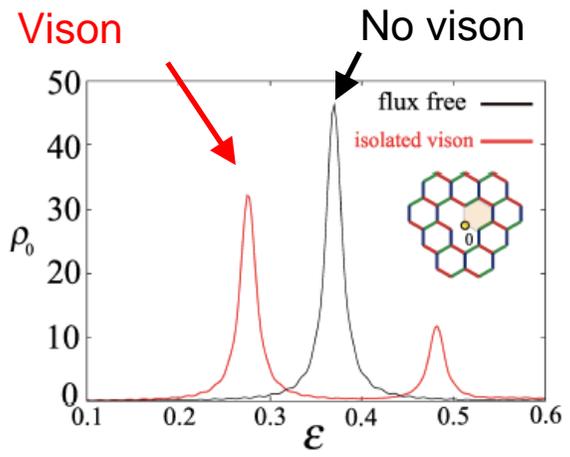
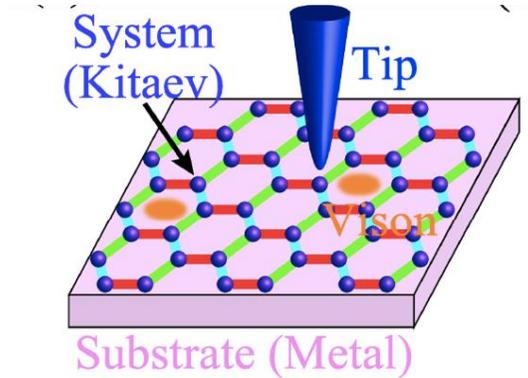
J. Feldmeier, *et al.*,
Phys. Rev. B **102**, 134426 (2020).

Spin gap



E. J. König *et al.*,
Phys Rev. Lett. **125**, 267206 (2020).

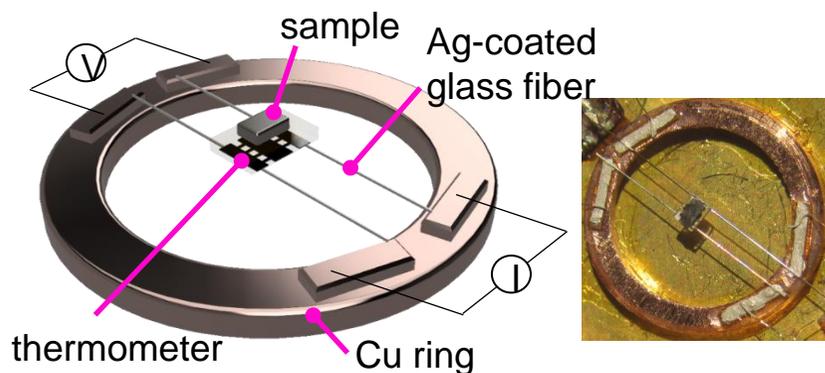
Non-abelian anyon



M. Udagawa, *et al.*,
Phys. Rev. Lett. **126**, 127201 (2021).

High-resolution specific heat measurements

Long-relaxation calorimetry

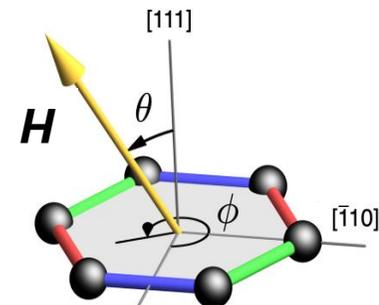
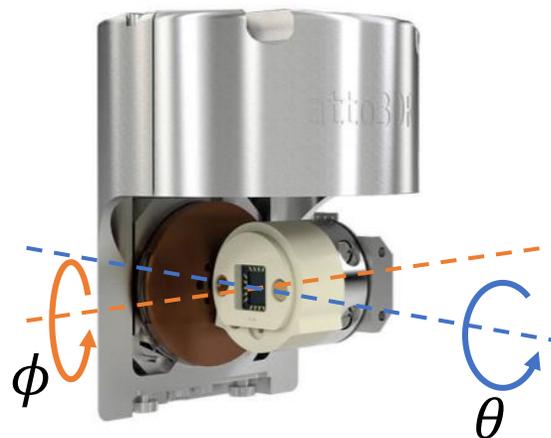


Our addenda ~ 100 pJ/K
Typical PPMS > 100000 pJ/K

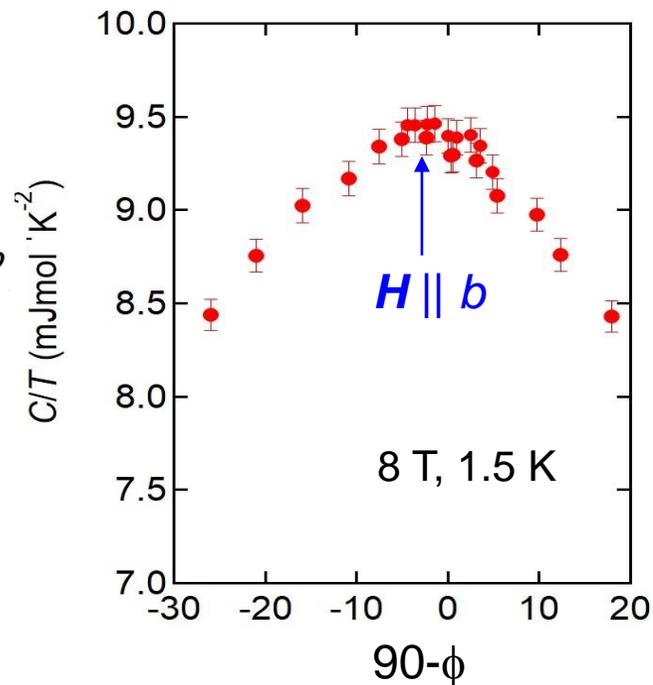
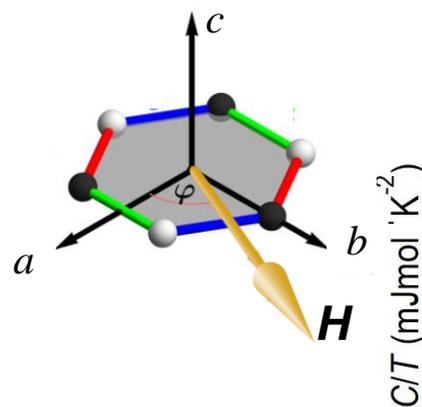
Precise determination of C for small single crystals (10 - $100 \mu\text{g}$) down to 250 mK.

Magnetic field direction can be controlled very precisely with respect to the crystal axis.

Two-axis rotator

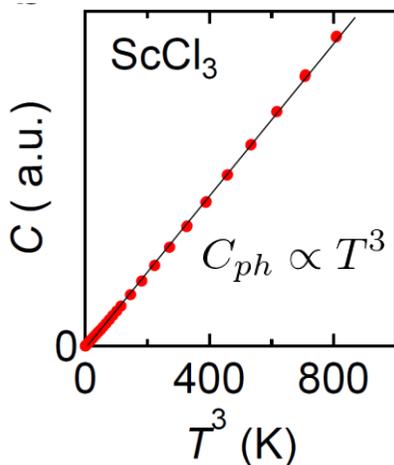
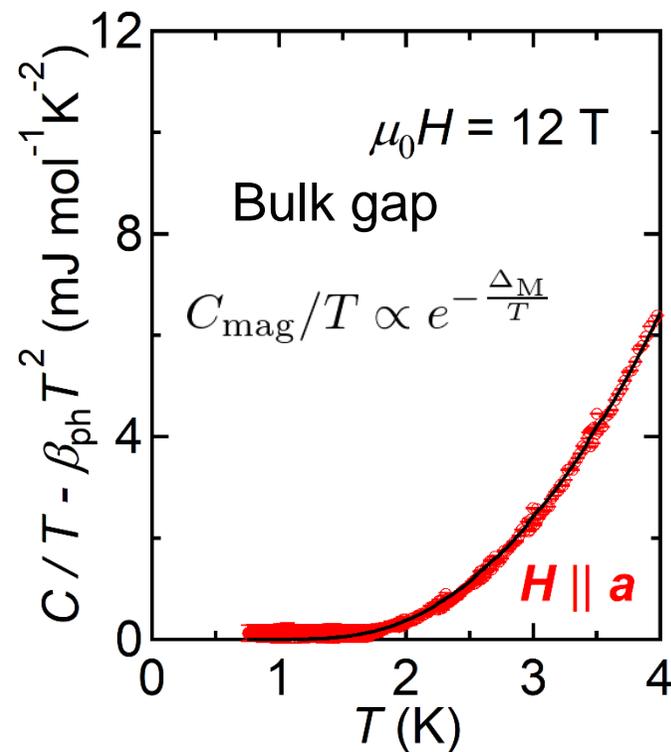
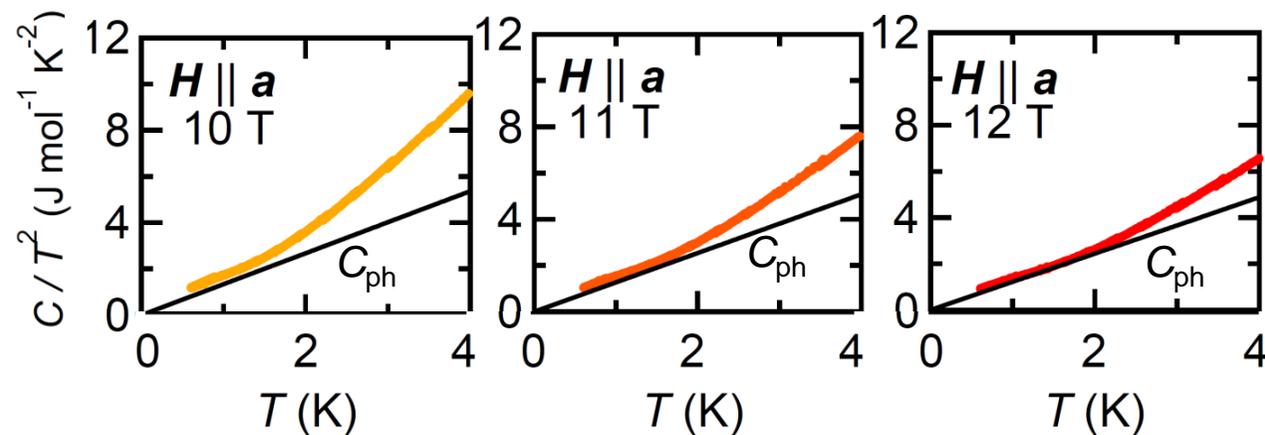


In-plane field rotation ($\theta=90^\circ$)

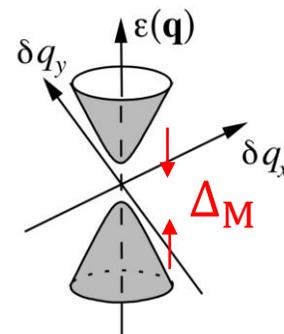


Majorana gap for $H//a$: specific heat in FIQD state

O. Tanaka *et al.* Nature Phys. **18**, 429 (2022).



Nonmagnetic
reference



$$\Delta_M \sim H^3$$

Consistent with Majorana gap

Reproducibility of thermal Hall effect

	sample	κ_{xy} plateau	$\kappa_{xy}^{2D} = K_0/2$	sign of κ_{xy}	Planar thermal Hall
Kyoto ^{1,2}	Bridgman <i>Tokyo Institute of Technology</i>	Yes	Yes	+60°+45° $\kappa_{xy} < 0$ -60°-45° $\kappa_{xy} > 0$ $H \parallel -a : \kappa_{xy} > 0$	Yes
ISSP Tokyo ³	Bridgman <i>Tokyo Institute of Technology</i>	Yes	Yes	+60° $\kappa_{xy} < 0$	
MPI Stuttgart ⁴	Bridgman <i>Tokyo Institute of Technology</i>	Yes	Yes	-70° $\kappa_{xy} > 0$	
Princeton ⁵	CVT <i>Oak Ridge</i>	No	No		Yes
Oak Ridge ⁶	CVT <i>Oak Ridge</i>	Yes	~75% of $K_0/2$	$H \parallel -a : \kappa_{xy} > 0$	Yes

Inconsistent with Kitaev QSL

Consistent with Kitaev QSL

1) Y. Kasahara, YM *et al.* Nature **559**, 227 (2018).

2) T. Yokoi, YM *et al.* Science **373**, 568 (2021).

3) M. Yamashita *et al.* PRB **102**, 220404(R) (2020).

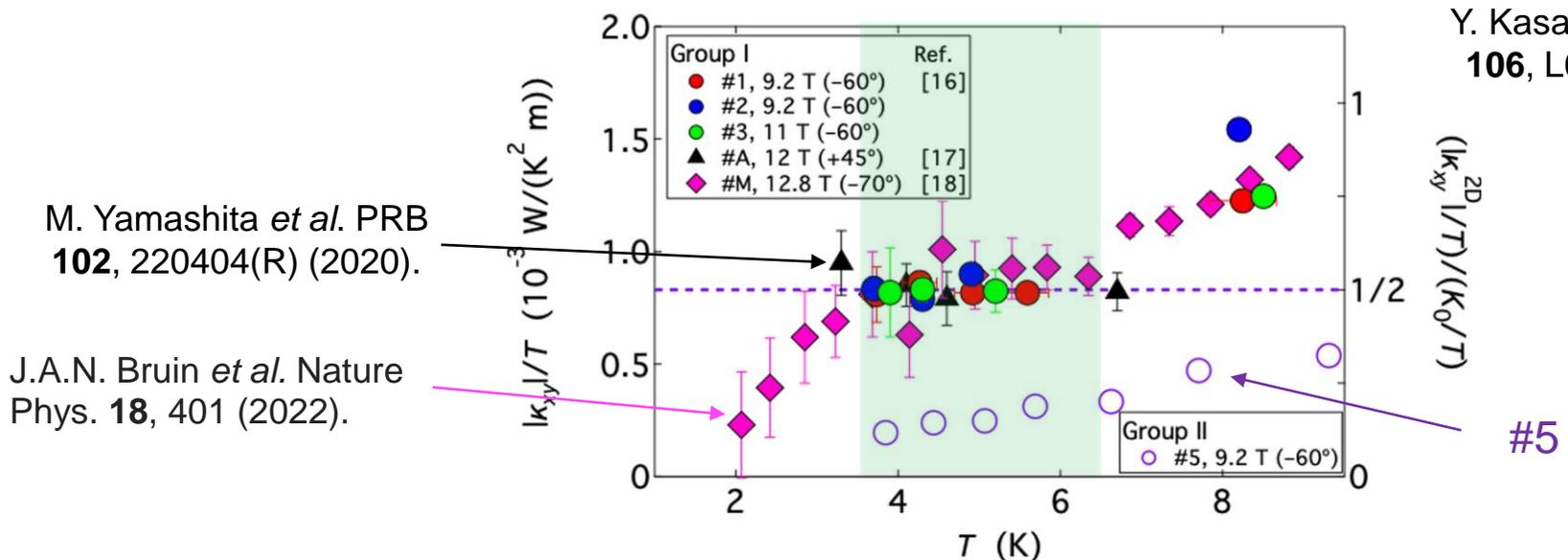
4) J.A.N. Bruin *et al.* Nature Phys. **18**, 401 (2022).

5) P. Czajka *et al.* Nature Phys. **17**, 915 (2021).

6) H. Zhang *et al.* arXiv:2303.03682.

Quantized and unquantized thermal Hall effect

Y. Kasahara, *et al.* PRB **106**, L060410 (2022).



For further confirmation of Majorana fermion, we need to clarify

1. Topology
 Chern number, Dirac cone, Majorana gap etc.
2. Reproducibility



Y. Vinkler-Aviv and A. Rosch, PRX **8**, 021032 (2018).
 M.Ye, *et al.*, PRL **121**, 147201 (2018).

Kitaev quantum spin liquid

Spin-1/2 on 2D honeycomb lattice with three inequivalent bonds

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

$$\mathcal{H} = -J_x \sum_{\langle ij \rangle_x} S_i^x S_j^x - J_y \sum_{\langle ij \rangle_y} S_i^y S_j^y - J_z \sum_{\langle ij \rangle_z} S_i^z S_j^z$$

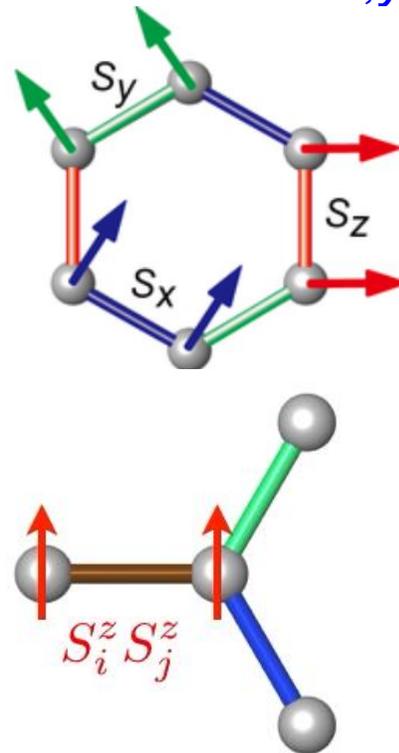
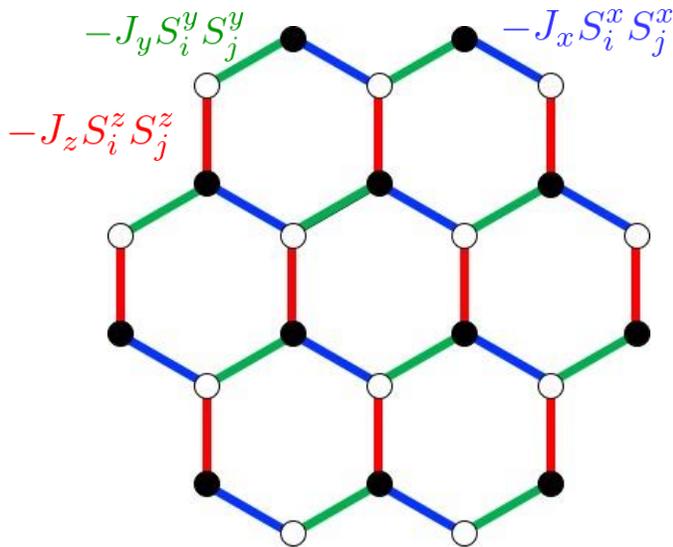
Kitaev interaction

Bond-dependent Ising-like

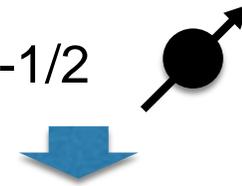
Each bond favors different directions of x, y and z.

➔ **Exchange frustration**

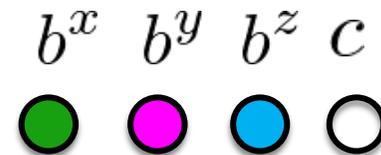
2D Honeycomb



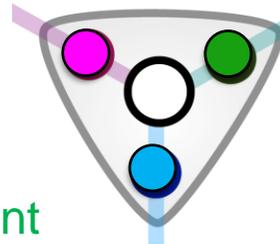
Spin-1/2



Majorana fermions

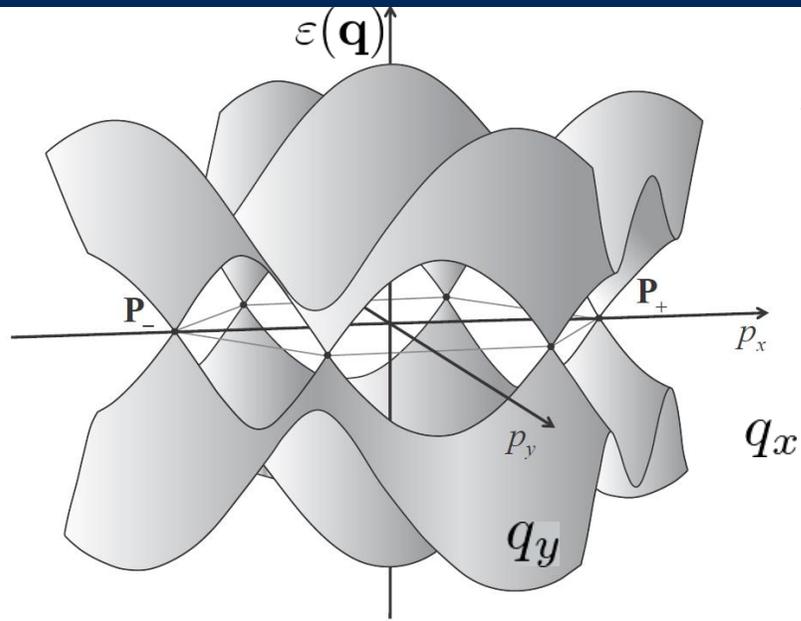


Localized Itinerant

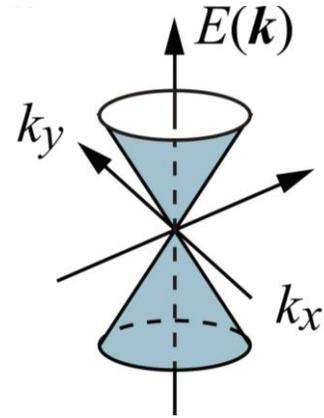


$$b^{\alpha\dagger} = b^\alpha \quad c^\dagger = c$$

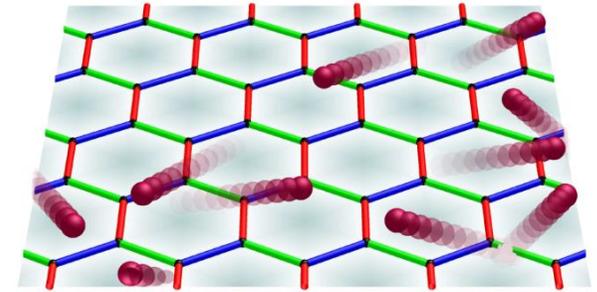
Energy dispersion of itinerant Majorana



$$H = 0$$

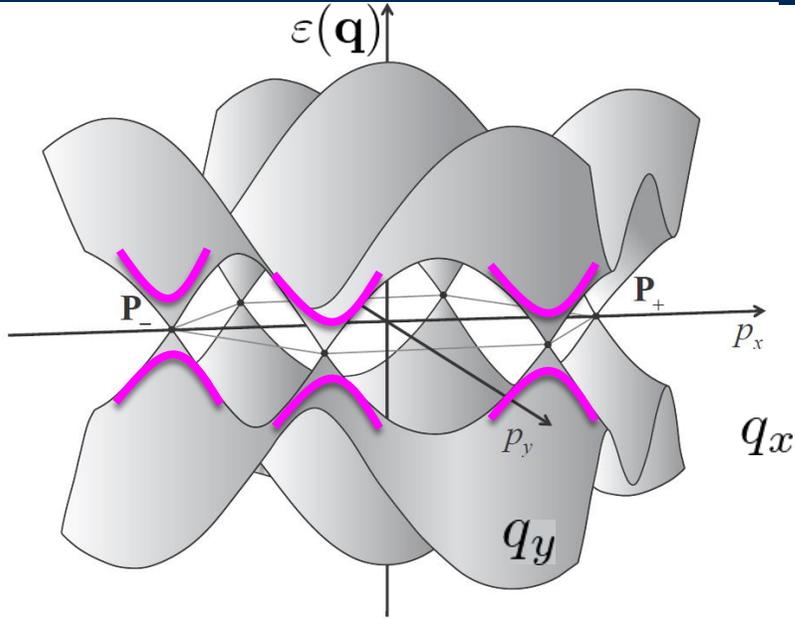


Itinerant Majorana

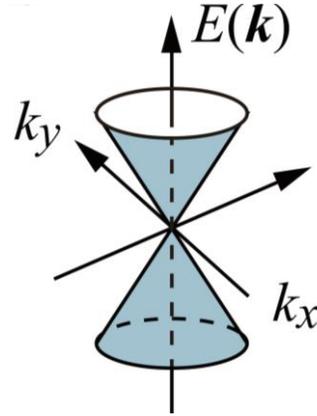


Majorana semimetal

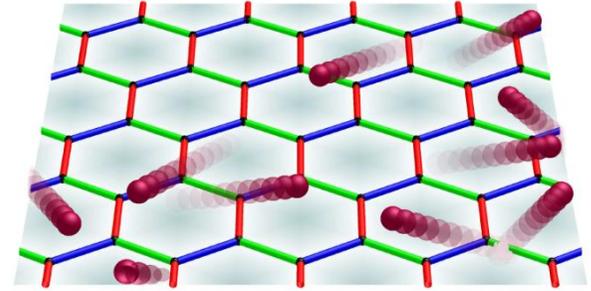
Energy dispersion of itinerant Majorana



$H = 0$



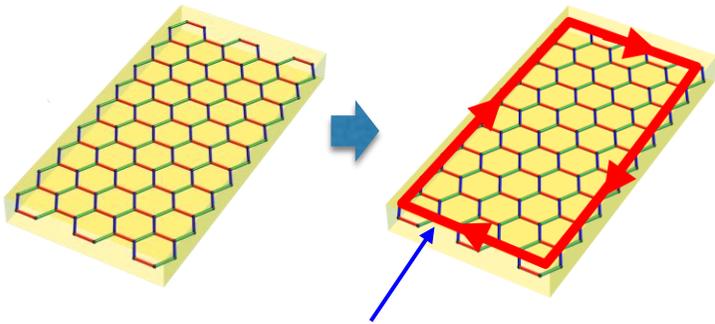
Itinerant Majorana



Majorana semimetal

$H = 0$

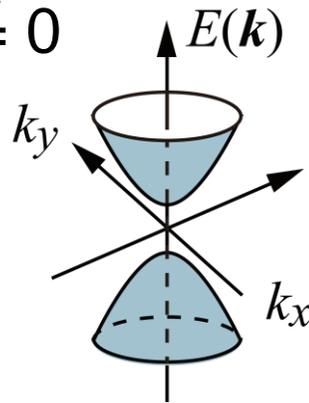
$H \neq 0$



Chiral Majorana edge current

One Majorana = "half" of a fermion

$H \neq 0$



Majorana Chern insulator

Chern number $C_h = \frac{\Delta}{|\Delta|} = \pm 1$

$$\kappa_{xy}^{2D} = \frac{1}{2} K_0$$

$$K_0 = \frac{\pi^2 k_B^2 T}{3h}$$

Quantum Thermal Conductance

Half integer thermal QHE

$$\ominus = \frac{1}{2} (\bullet + \circ)$$