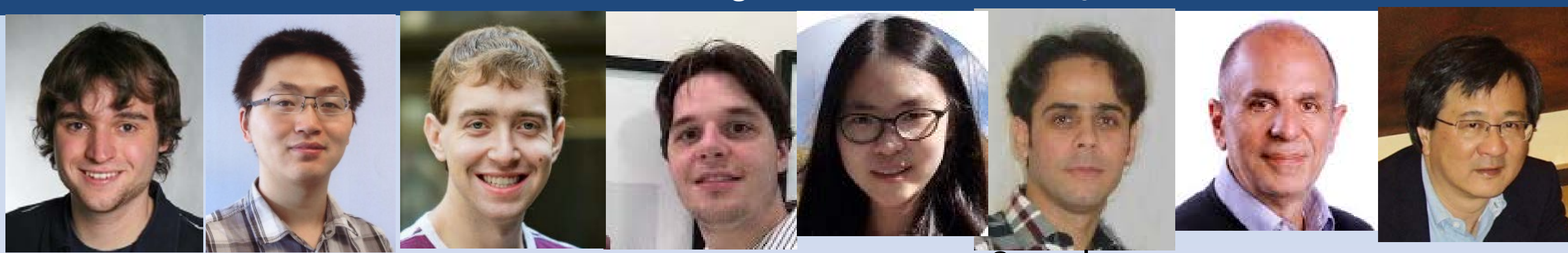
Thermal Hall Effect from Neutral Spin Excitations in Frustrated Quantum Magnets

N. P. Ong, *Princeton University*



Max Hirschberger | Tong Gao | Peter Czajka | Jason Krizan | Ruidan Zhong | Seyed Koohpayeh JHU | Cava | NPO

1. Thermal Hall effect as a transport probe of spin liquids
2. The pyrochlores $Tb_2Ti_2O_7$ and $Yb_2Ti_2O_7$
3. A new Co-based triangular lattice frustrated magnet

Work at Princeton supported by Moore Foundation, DOE, ARO, NSF

Resonating Valence Bond State (Anderson, 1971, 1987)

Anderson

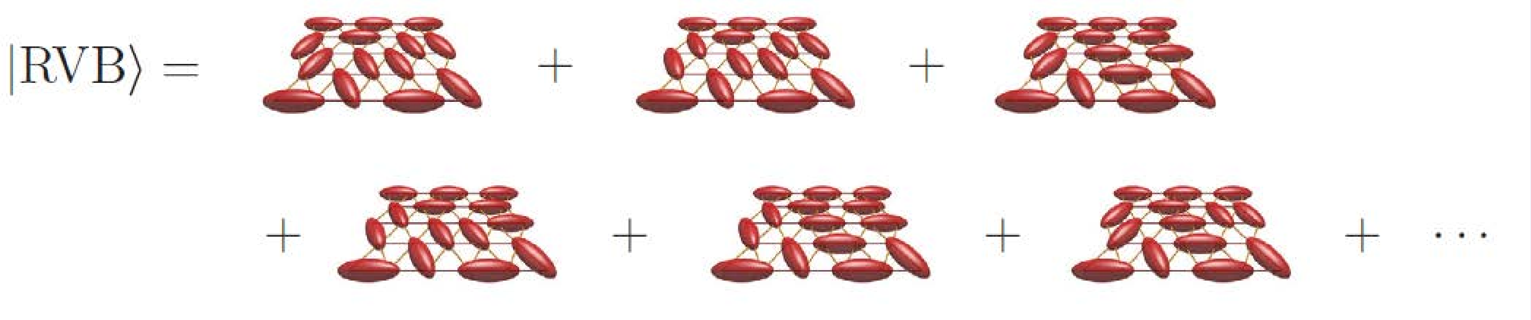
In 2D triangular lattice, strong quantum fluctuations can lead to disordered spin-liquid state with holon and spinon excitations

Superposition of all possible singlets

$$|RVB\rangle = \sum_{ij} (\uparrow_i \downarrow_j - \downarrow_i \uparrow_j)/\sqrt{2}$$

QSL is a quantum state with anomalously high entanglement and massive superposition

Savary and Balents, *Rep. Prog. Phys*. (2017)



Topological order

A phase of quantum matter outside the purview of the Landau paradigm

1) Highly entangled ground state that cannot be described by
Product State $\Psi = \sigma_1 \sigma_2 \sigma_3 \sigma_4 \sigma_{51} \ldots$

2) Should have topological excitations that cannot be reduced to local operators
(visons, spinons, anyons, Majoranas, fauxtons, …)

Examples

i) Haldane *S* = 1 spin chains ($CsNiCl_3$)

ii) FQHE

iii) Gapped spin liquids
- a) Kitaev Toric Code model
- b) Kitaev hexagonal spin model
- c) Z_2 spin liquid

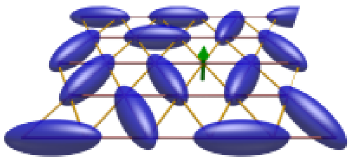
Recent reviews on Quantum Spin Liquids

i) Savary and Balents, Rep. Prog. Phys. 2017

ii) Zhou, Kanoda and Ng, RMP 2017

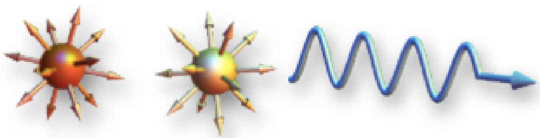
Classes of QSLs

- Topological QSLs



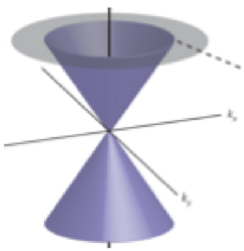
projected superconductor

- U(1) QSL



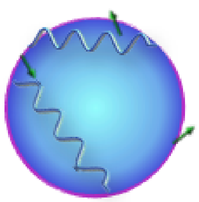
projected 3d band insulator

- Dirac QSLs



projected graphene

- Spinon Fermi surface



projected metal

Quantum Spin Liquid Candidates

Triangular Lattice

κ-(BEDT-TTF)2Cu2(CN)3

EtMe3Sb[Pd(dmit)2]2

CsCuCl4

Ba3CuSb2O9

YbMgGaO4

Sc4Ga4CuO7

1T-TaS2

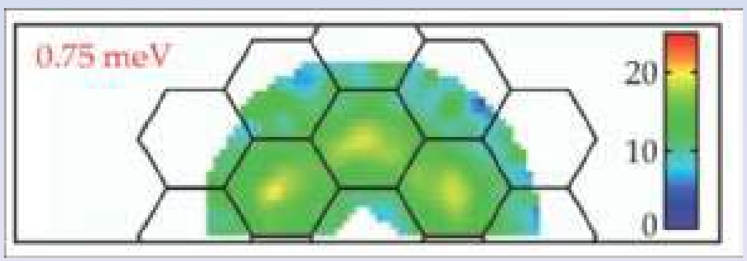
Kagome Lattice
HyperKagome

α-RuCl3

ZnCu3(OH)6Cl2

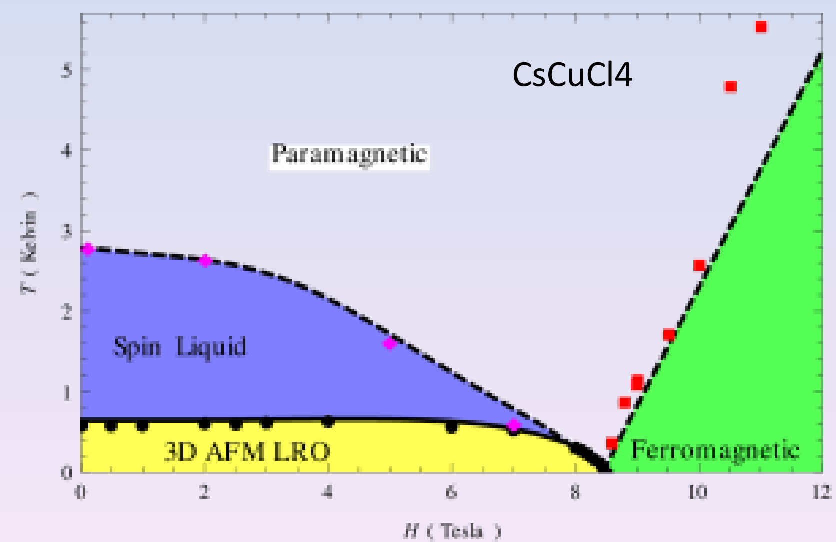
Na4Ir3O8

ZnCu3(OH)6Cl2

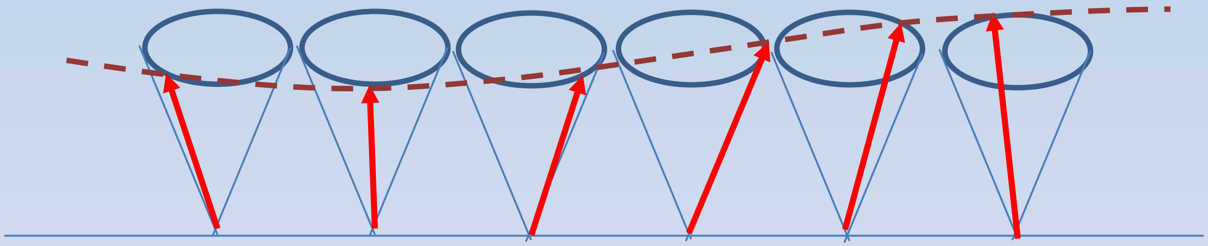


Neutron diff.: gapless
NMR: gapful

Coldea R. et.al. RPL (2001)
Han T. et.al. Nature (2010)



Spin excitations of a (anti-) ferromagnet are magnons (spin waves)



In quantum frustrated magnets, what are the excitations?
Do they transport energy?

Major challenge: phonon conduction usually dominant

Materials

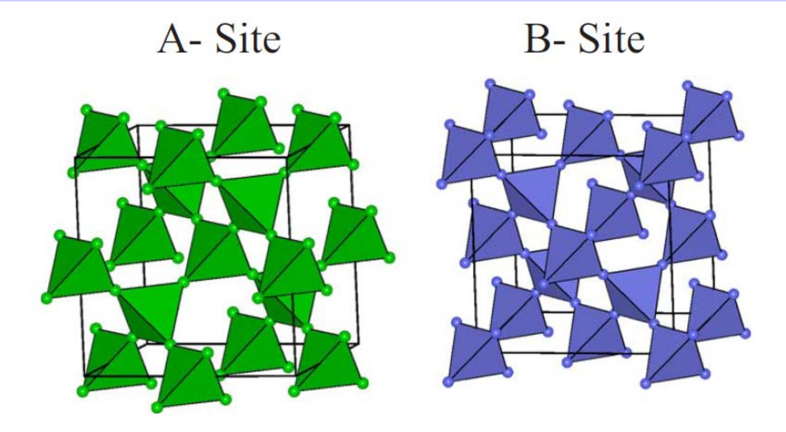
Experiments on a frustrated quantum pyrochlore magnet (spin liquid candidate)
And an ordered Kagome magnet

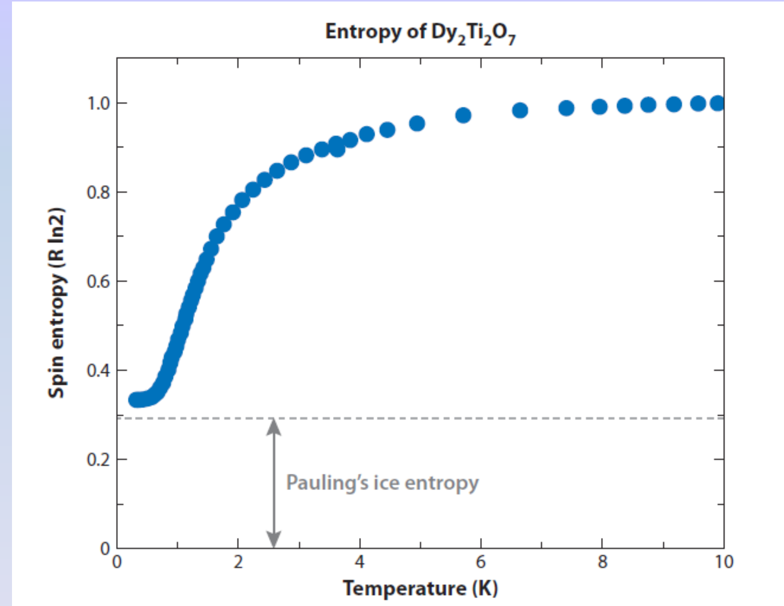
1) Pyrochlore $Tb_2Ti_2O_7$
   High-temp suscep yields Curie-Weiss MFT temp of 19 K but
   fails to order down to 50 mK.
   Ground state "Quantum spin ice", may harbor spin liquid

2) Pyrochlore $Yb_2Ti_2O_7$
   Orders at 250 mK

3) A new candidate $Na_2BaCo(PO_4)_2$

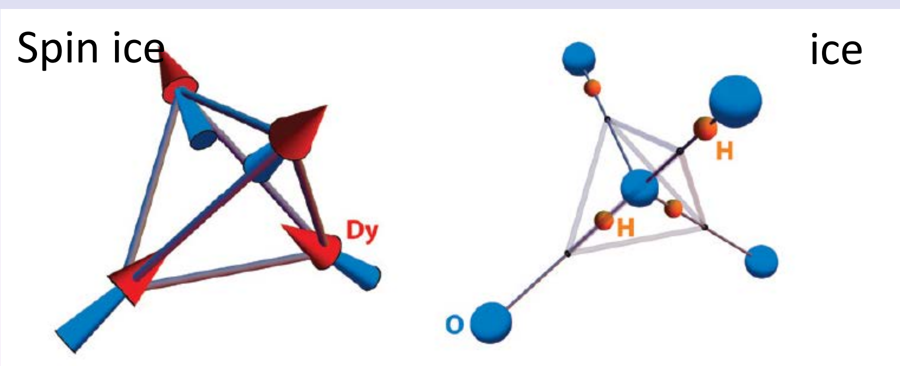
Pyrochlores, spin-ice systems





Ramirez, et al. Nature 1999

Two-in, two-out config.



Castelnovo, Moessner, Sondhi
Annu. Rev. Cond. Mat. 2012

Classical Spin ice

$Dy_2Ti_2O_7$, $Ho_2Ti_2O_7$

Quantum spin ice (no trace of 2-in/2-out)

$Tb_2Ti_2O_7$, $Yb_2Ti_2O_7$

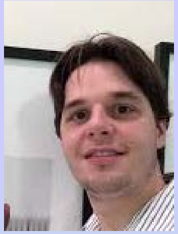
FRUSTRATED MAGNETISM

Science (2015)

Large thermal Hall conductivity of neutral spin excitations in a frustrated quantum magnet

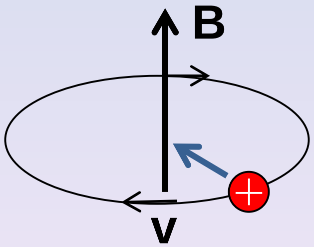
Max Hirschberger,[1] Jason W. Krizan,[2] R. J. Cava,[2] N. P. Ong[1]*

Hirschberger

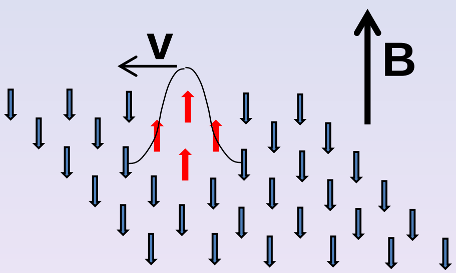


Krizan

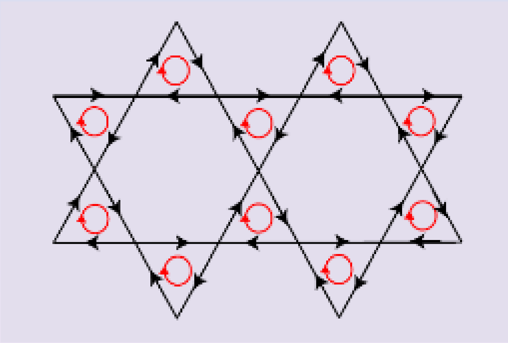
Can charge-neutral spin excitations display a Hall effect?



Charged excitation

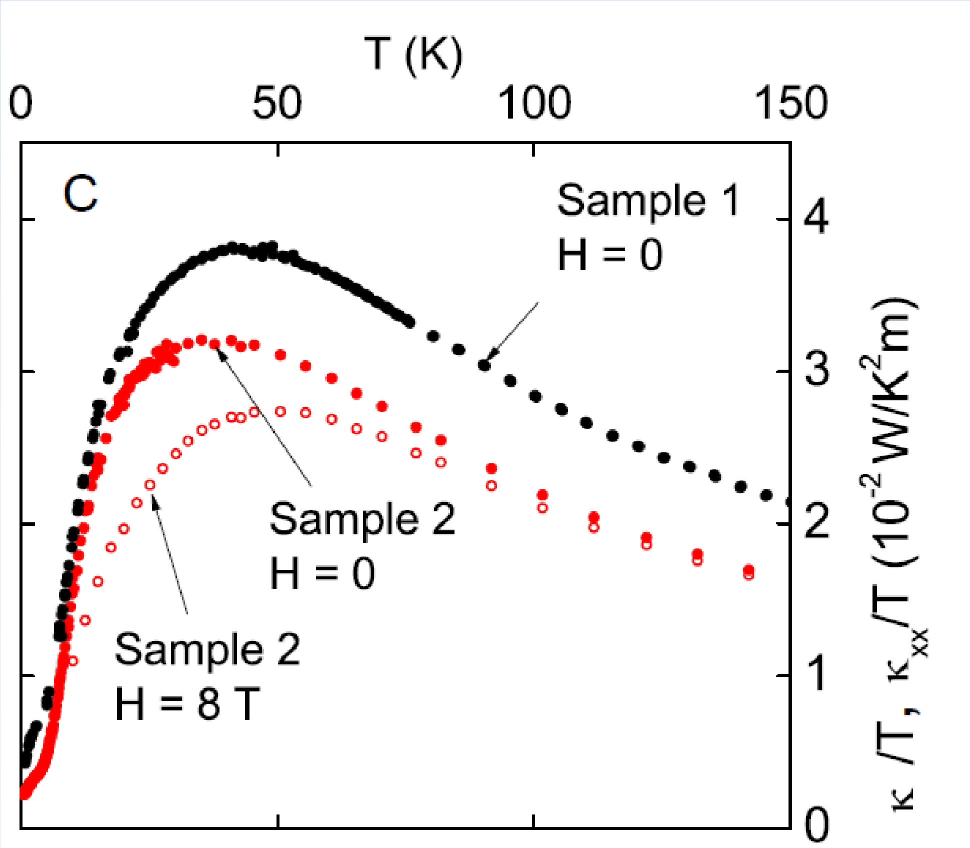


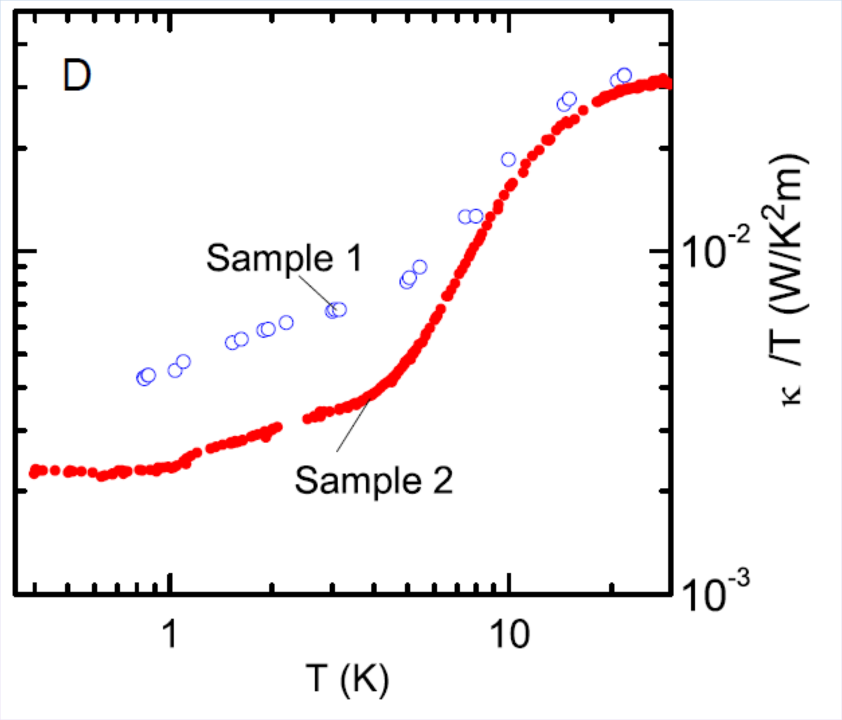
Wave packet of spin exc.
In ferromagnet



Chiral antiferromagnet

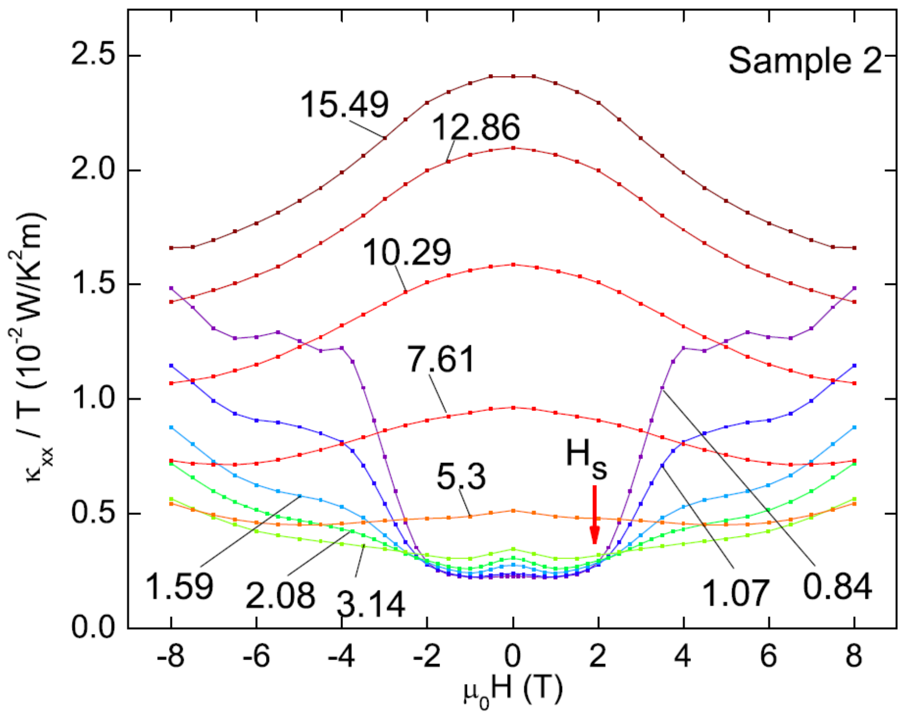
I) Thermal conductivity vs temp (B=0) in $Tb_2Ti_2O_7$

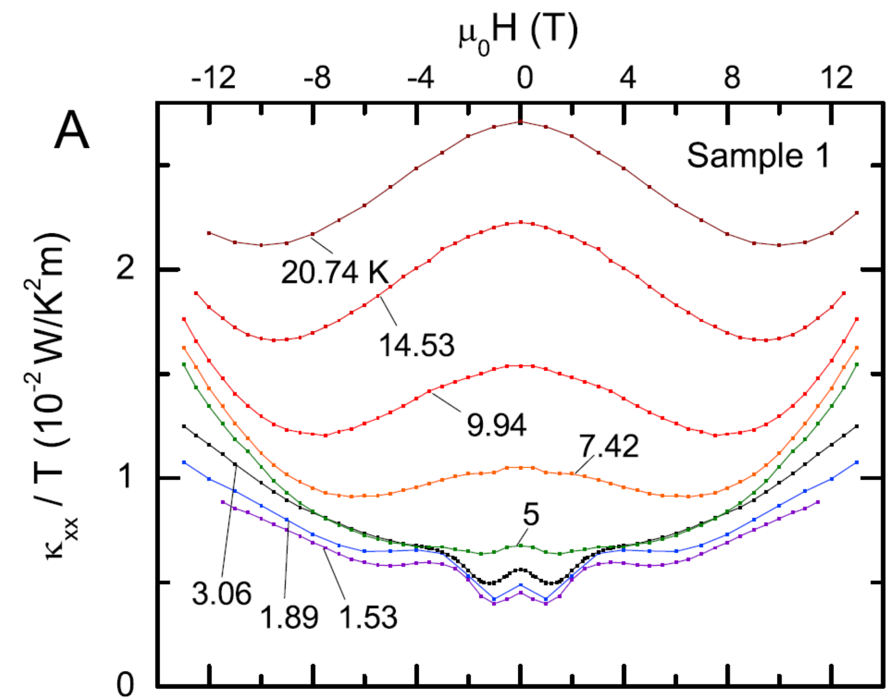




A very poor thermal conductor below 5 K

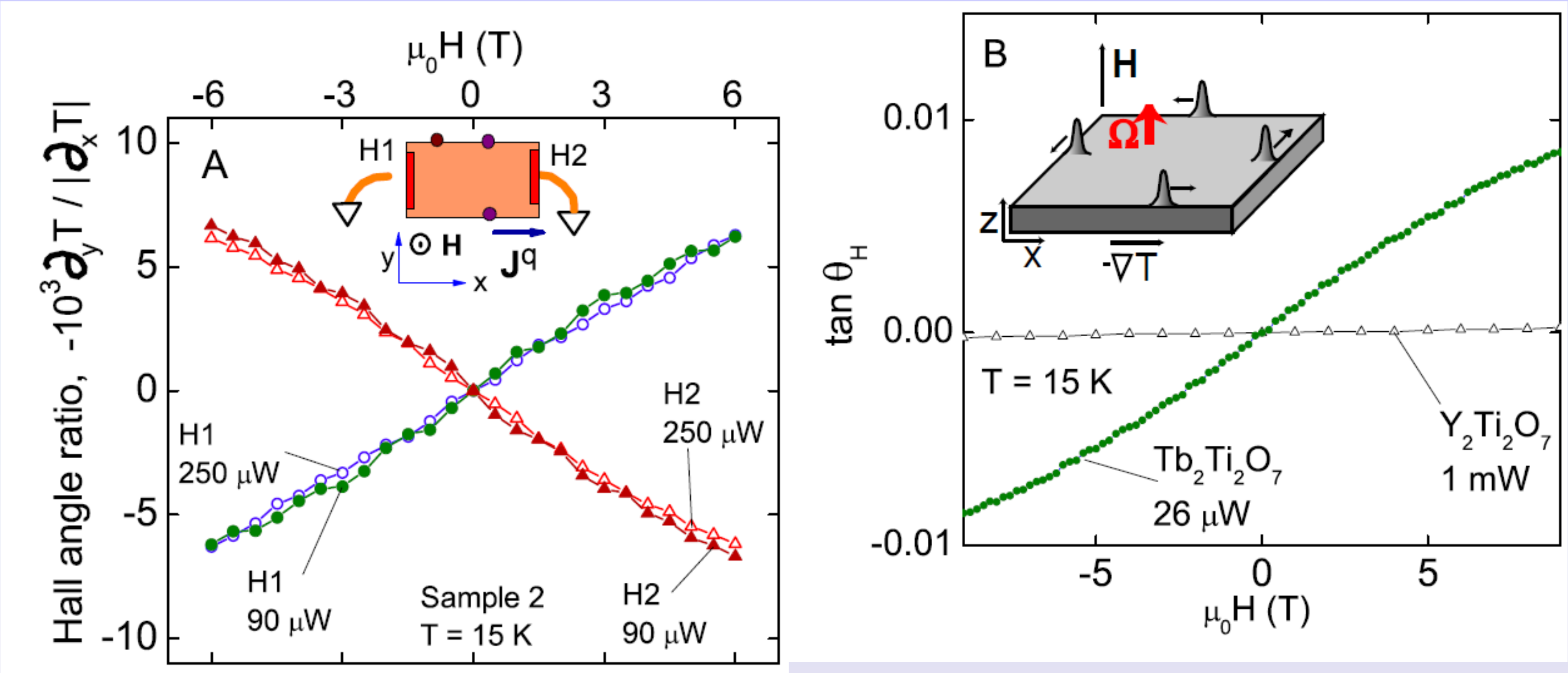
Magneto-thermal conductance T< 20 K





Large contribution to thermal current from spin excitations
Dominant below ~3 K
Very large field effect
Metamagnetic transition at H_s = 2 T leads to step-like increase in K_{xx}

Hall effect in a neutral current?? Experimental checks



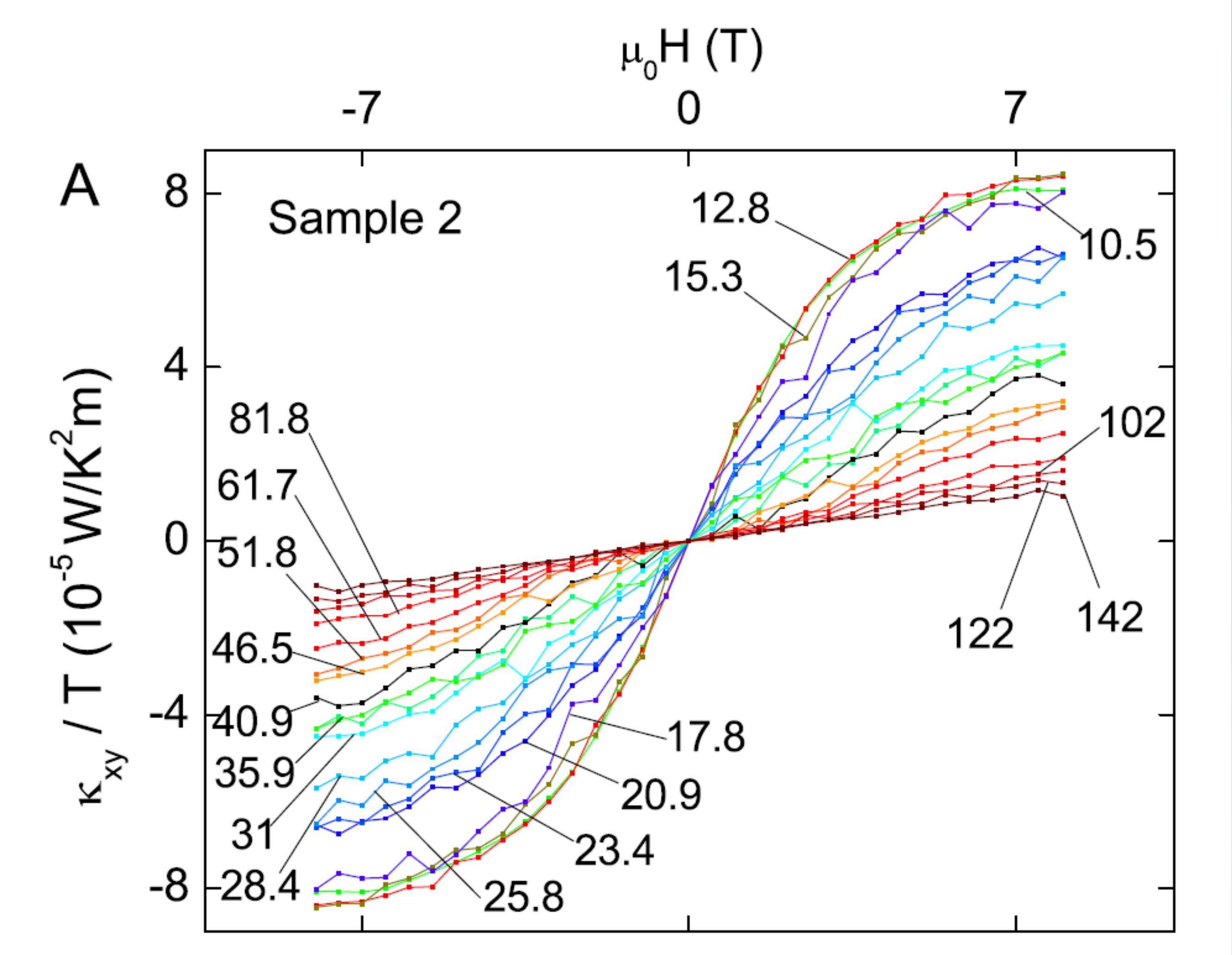
Checks

Hall signal reverses when gradient (-∇*T*) is reversed in same **B**

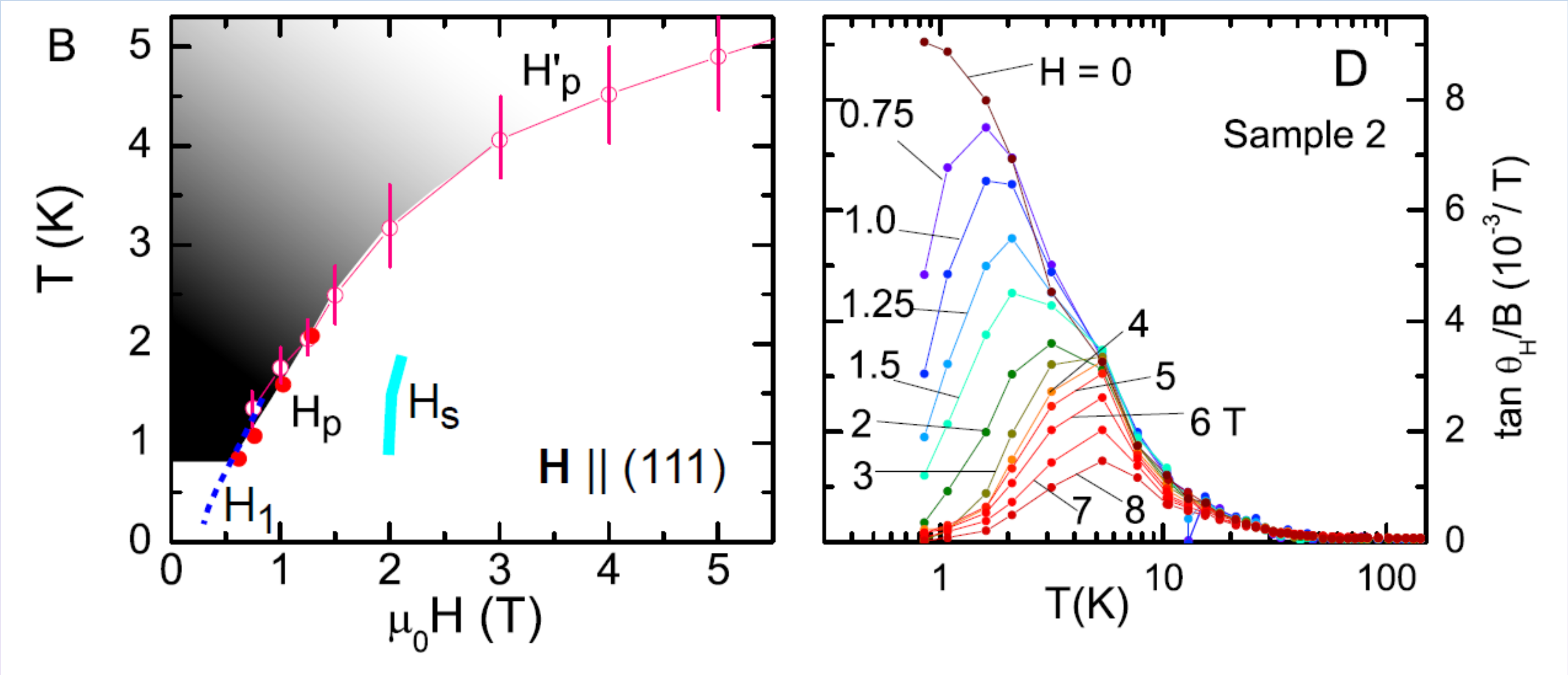
Hall signal scales linearly with gradient strength

Hall signal is 1000 x larger than in nonmag analog $Y_2Ti_2O_7$

Thermal Hall conductivity 10 < *T* < 140 K



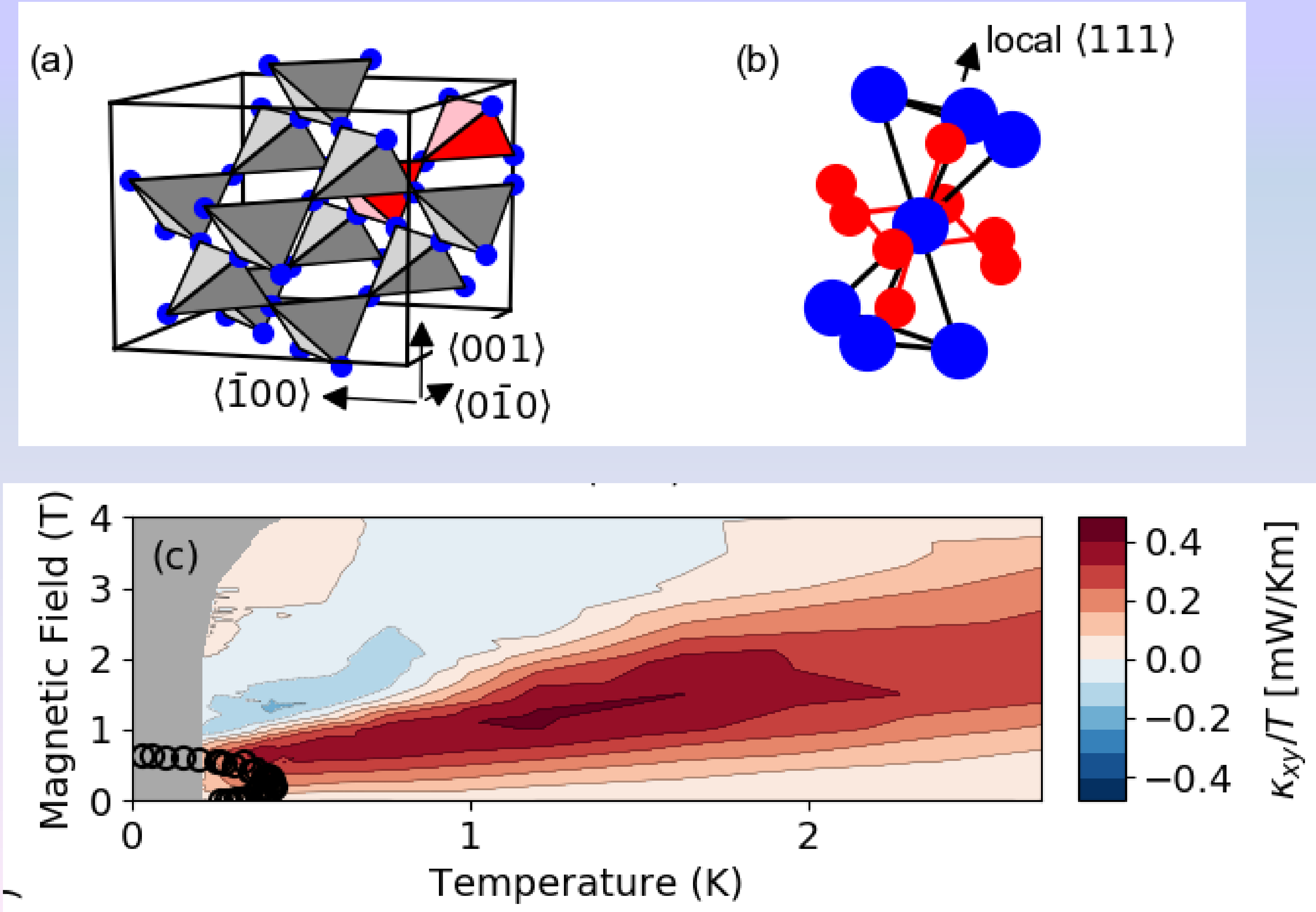
Phase diagram of large Hall state in the H-T plane



Extent of large-Hall response state in *T-H* plane (shaded).

Hall response is strongly suppressed in field-induced metamagnetic state ($H>H_s$)

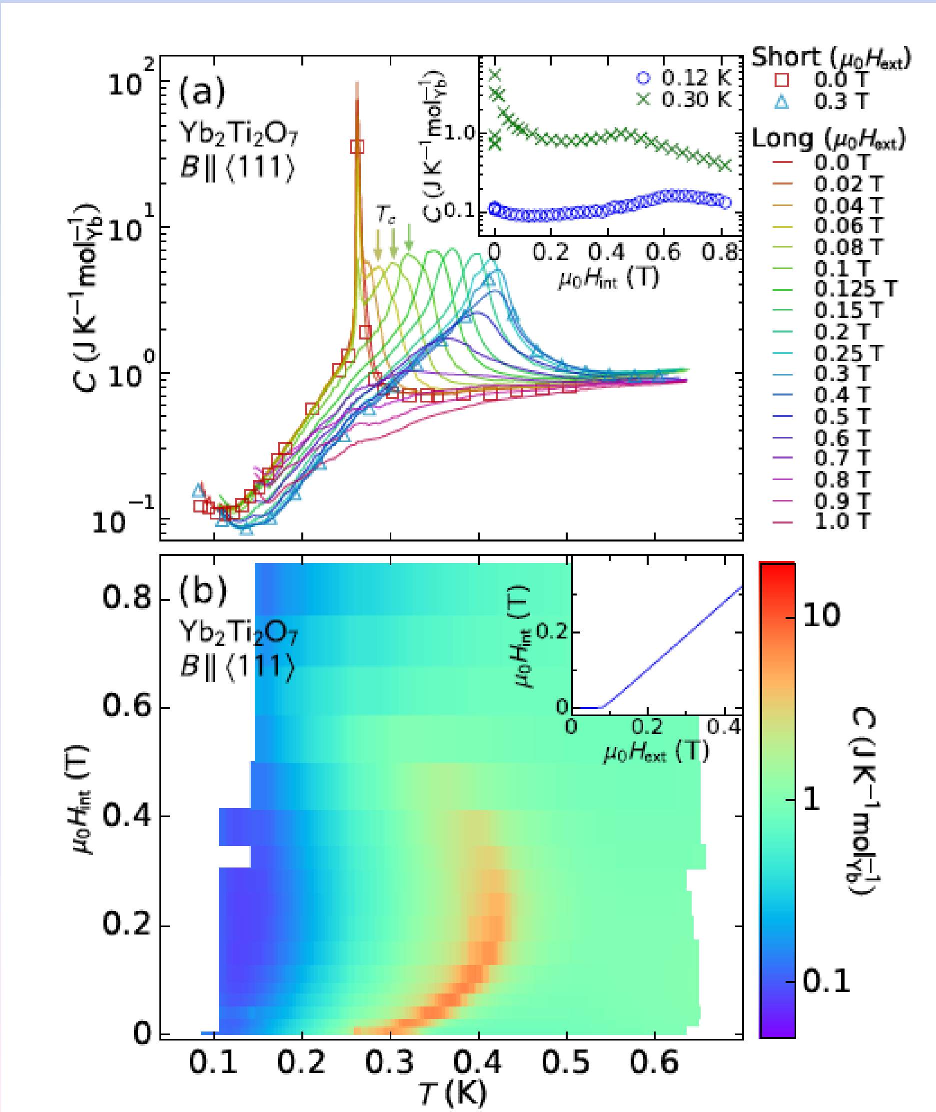
K_{xy} in a second pyrochlore $Yb_2Ti_2O_7$

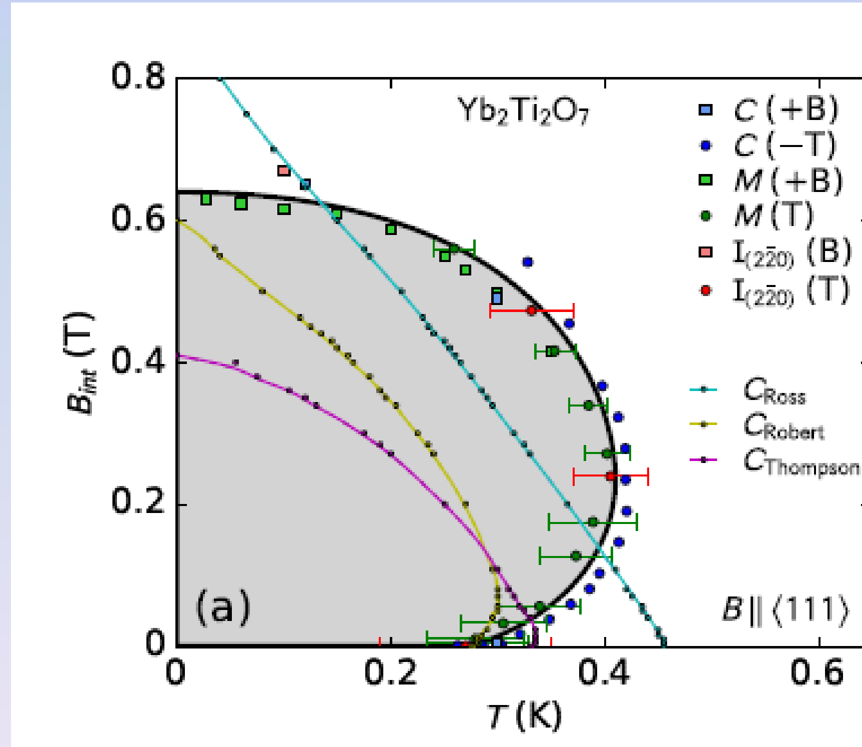

(a)
⟨001⟩
⟨1̄00⟩
⟨01̄0⟩
(b)
local ⟨111⟩
(c)
Magnetic Field (T)
Temperature (K)
κ_{xy}/T [mW/Km]
0.4
0.2
0.0
−0.2
−0.4

Reentrant Phase Diagram of $Yb_2Ti_2O_7$ in ⟨111⟩ Magnetic Field

JHU

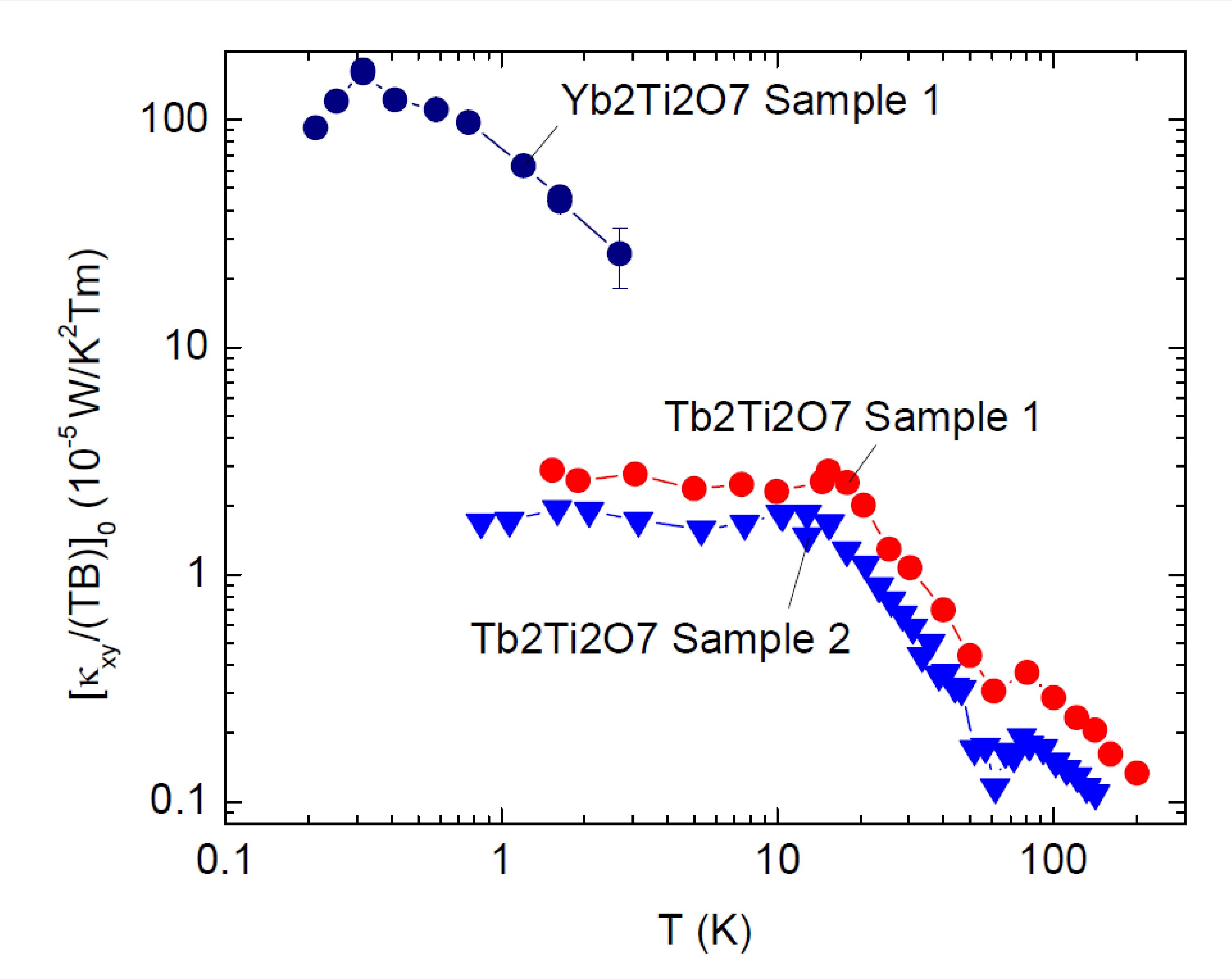
A. Scheie,[1,2] J. Kindervater,[1,2] S. Säubert,[3,4] C. Duvinage,[3] C. Pfleiderer,[3] H. J. Changlani,[1,2] S. Zhang,[1,2] L. Harriger,[5] K. Arpino,[6,2] S.M. Koohpayeh,[1,2] O. Tchernyshyov,[1,2] and C. Broholm[1,2,5,7]

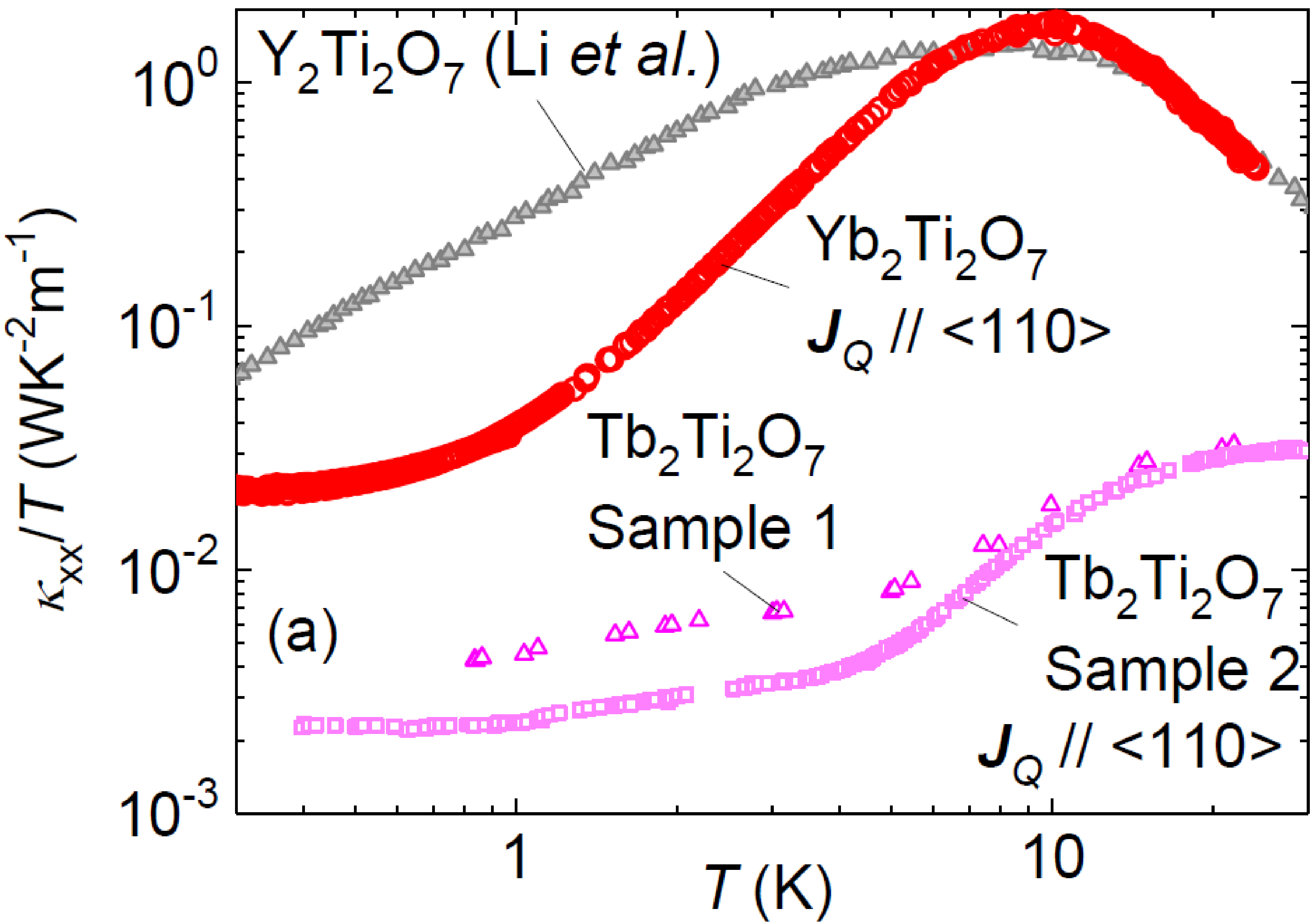




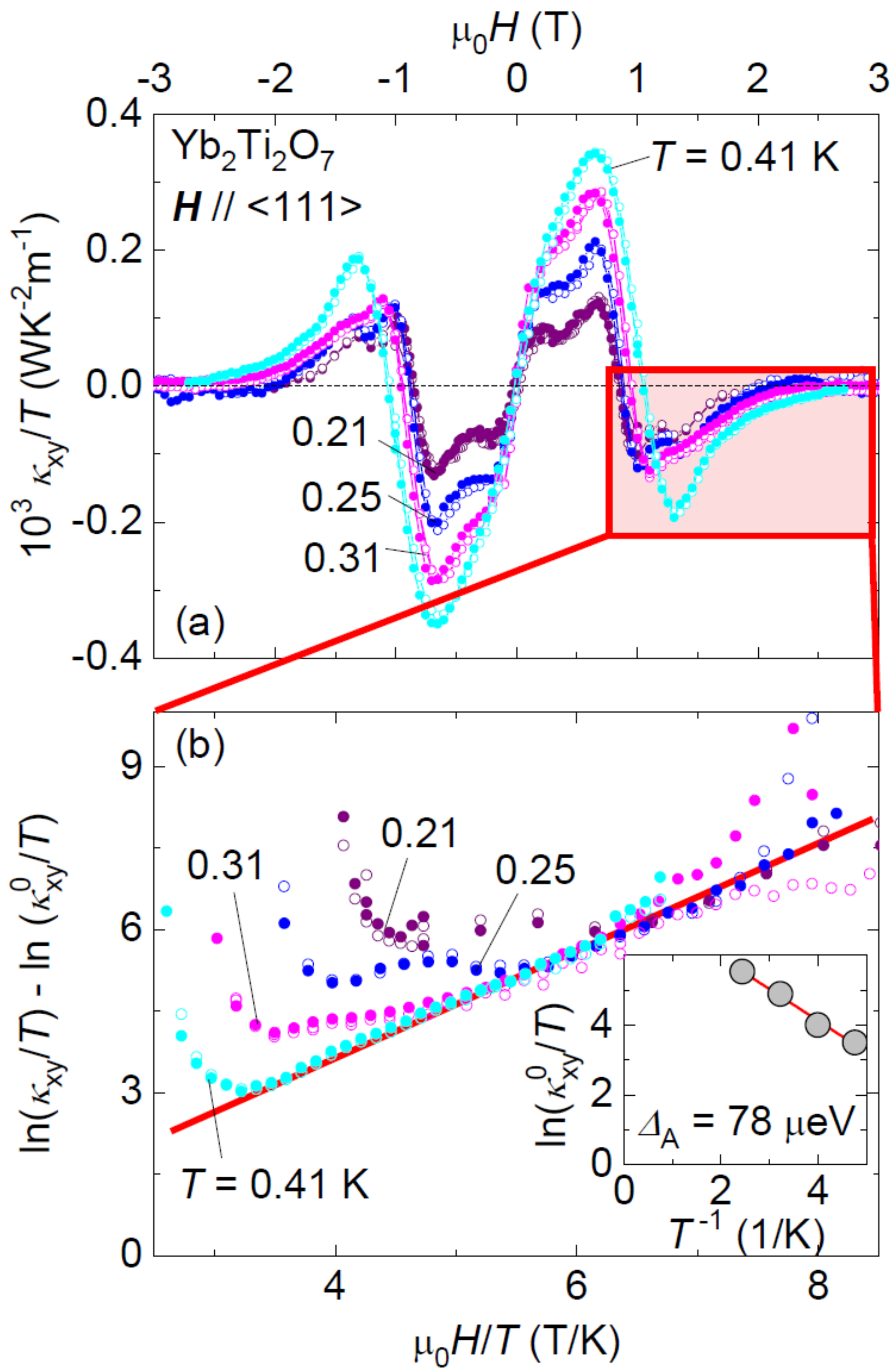
Weak-field thermal Hall response in $Tb_2Ti_2O_7$ and $Yb_2Ti_2O_7$

Hirschberger, Koohpayeh, Wang, NPO





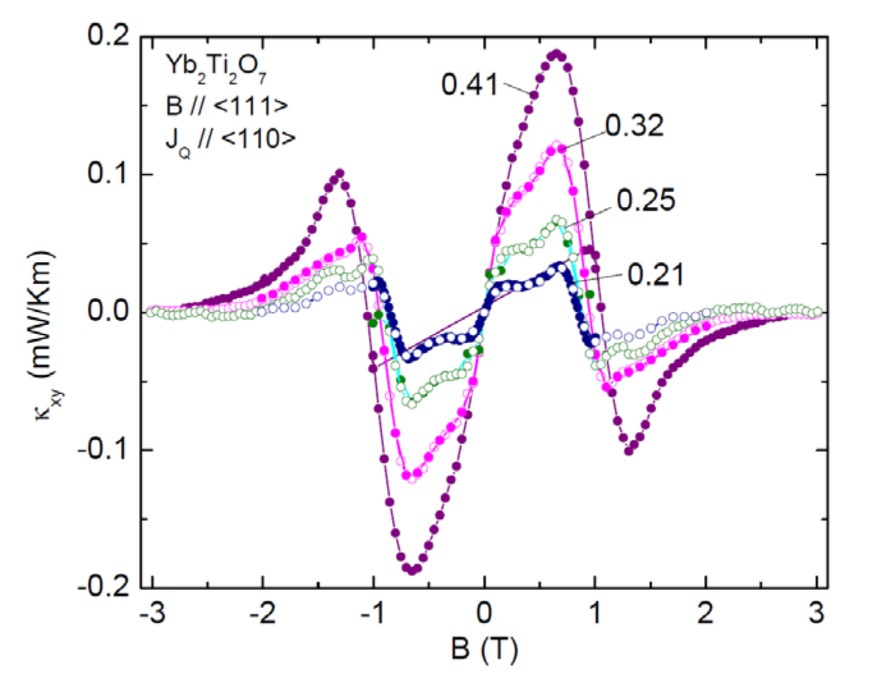
$Y_2Ti_2O_7$ (Li *et al.*)
$Yb_2Ti_2O_7$
J_Q // <110>
$Tb_2Ti_2O_7$
Sample 1
$Tb_2Ti_2O_7$
Sample 2
J_Q // <110>
(a)
10^0
10^{-1}
10^{-2}
10^{-3}
1
10
κ_{xx}/T ($WK^{-2}m^{-1}$)
T (K)

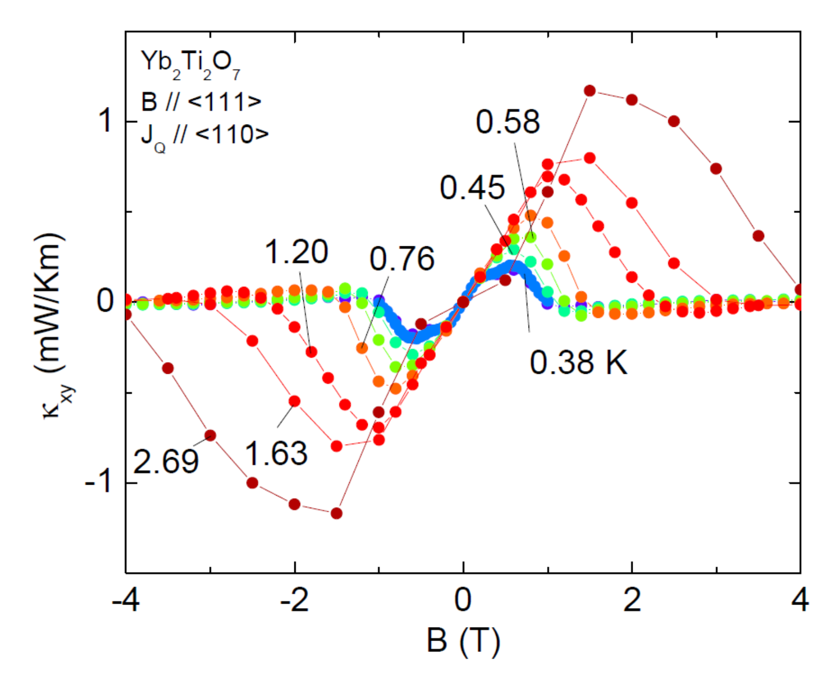


μ_0H (T)
-3
-2
-1
0
1
2
3
0.4
0.2
0.0
-0.2
-0.4
$10^3\ \kappa_{xy}/T\ (WK^{-2}m^{-1})$
$Yb_2Ti_2O_7$
H // <111>
T = 0.41 K
0.21
0.25
0.31
(a)
(b)
9
6
3
0
$\ln(\kappa_{xy}/T) - \ln(\kappa^0_{xy}/T)$
0.31
0.21
0.25
T = 0.41 K
4
6
8
μ_0H/T (T/K)
$\ln(\kappa^0_{xy}/T)$
4
2
0
0
2
4
T^{-1} (1/K)
Δ_A = 78 μeV

Distinctive field profile of K_{xy} in $Yb_2Ti_2O_7$

Hirschberger, Koohpayeh, Wang, NPO



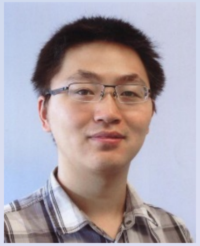


K_{xy} is large in frustrated state (weak B).
Suppressed when magnons appear above 2 T.

Signature of excitations in quantum disordered state?

Thermal Hall Effect in $Na_2BaCo(PO_4)_2$ a new candidate Quantum Spin Liquid

Tong Gao, Ruidan Zhong, R. J. Cava, N. P. Ong



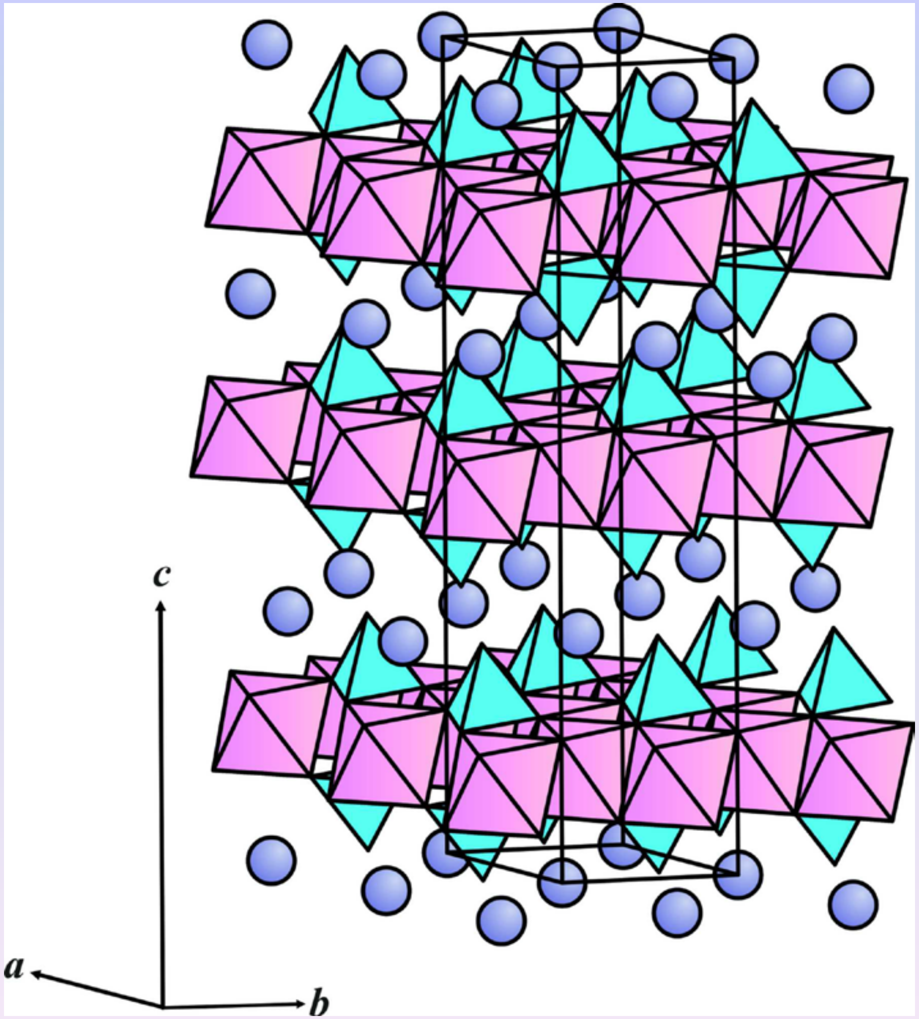
Tong Gao

Ruidan Zhong

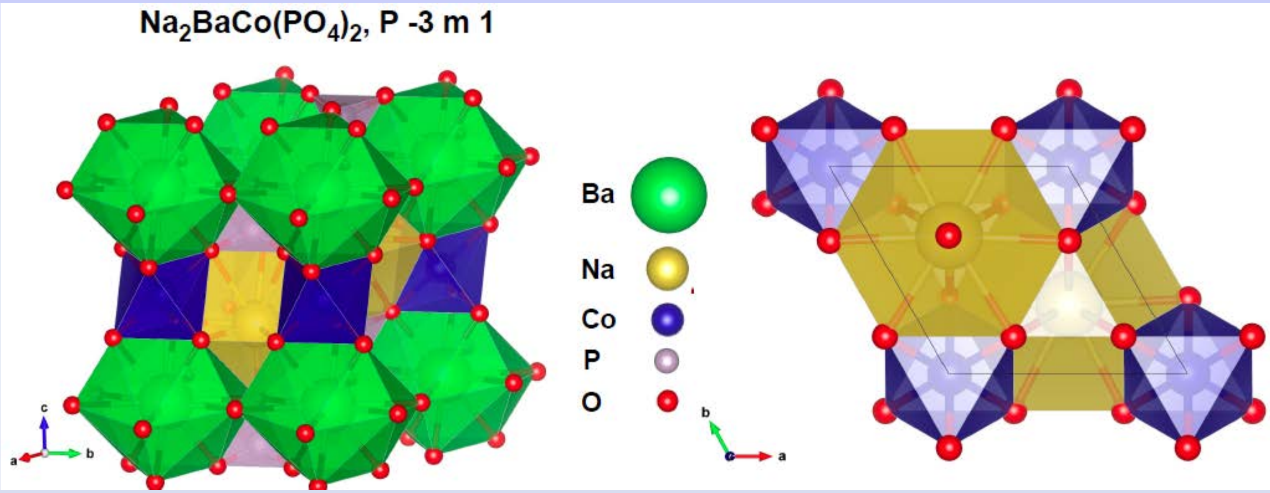
Cava

NPO

$Na_2BaCo(PO_4)_2$



$Na_2BaCo(PO_4)_2$



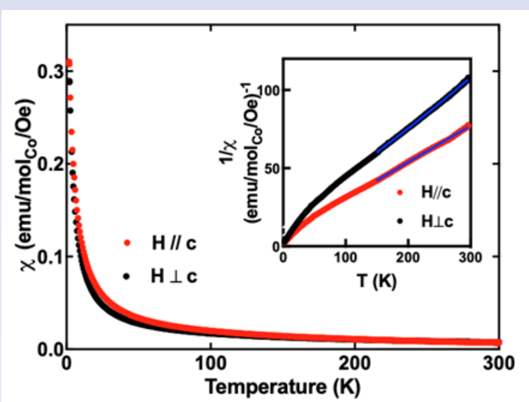
In plane:

Co – O – O – Co

Out of plane:

Co – O – O – O – Co

Effective 2-D triangular lattice



$\Theta_{CW} \sim -32\text{K}$

$J \sim 22\text{K}$

No ordering measured to 0.3K

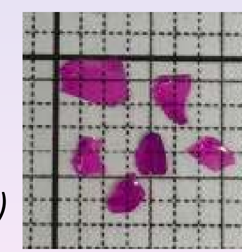
$f = \frac{|\Theta_{CW}|}{T_N} > 100$

No Site-mixing or other disorders

Co in isotropic lattice environment

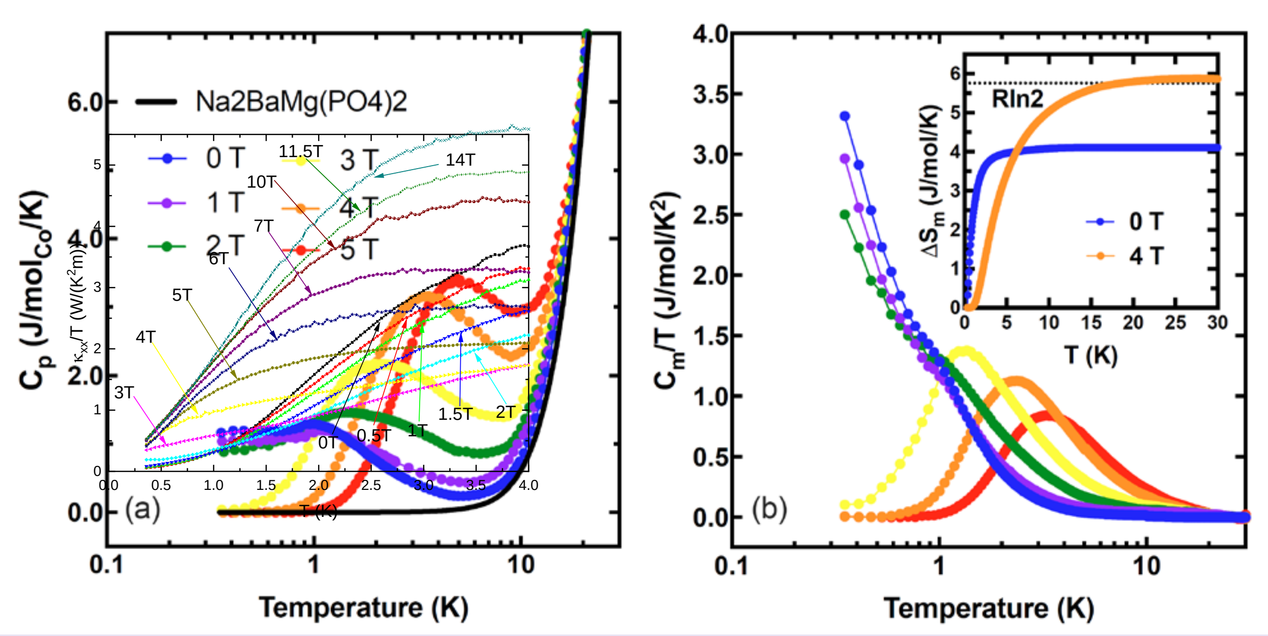
Perfect Candidate for geometrical frustration

Zhong R., et al. *Under review (2019)*



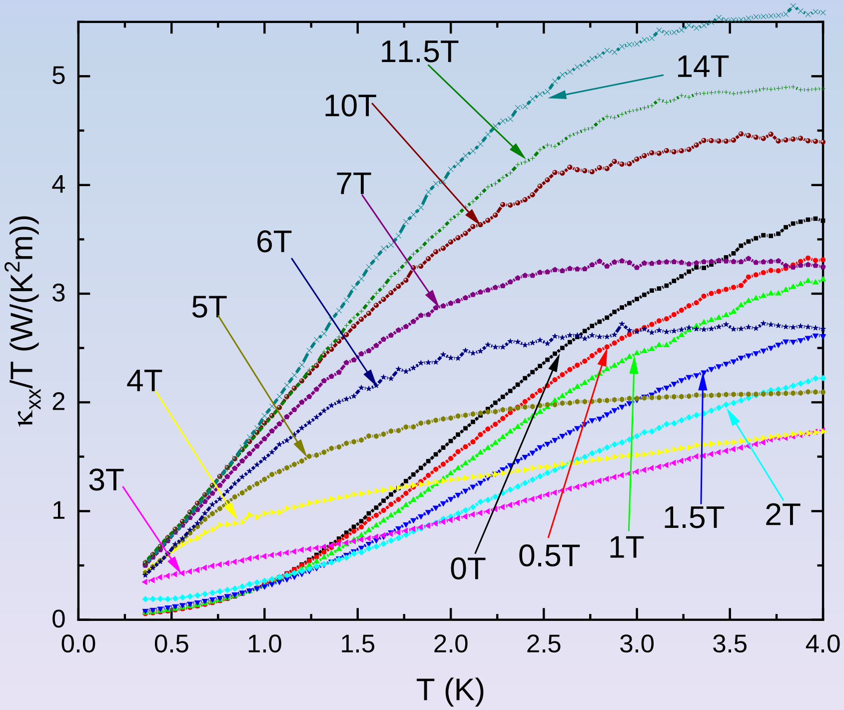
Magnetic Specific Heat of Na2BaCo(PO4)2

- No ordering features above 0.3K
- large heat capacity below 1K with small temperature dependency at 0T
- With >2T magnetic field, heat capacity vanishes quickly with temperature, indicating phase transition
- Huge magnetic entropy

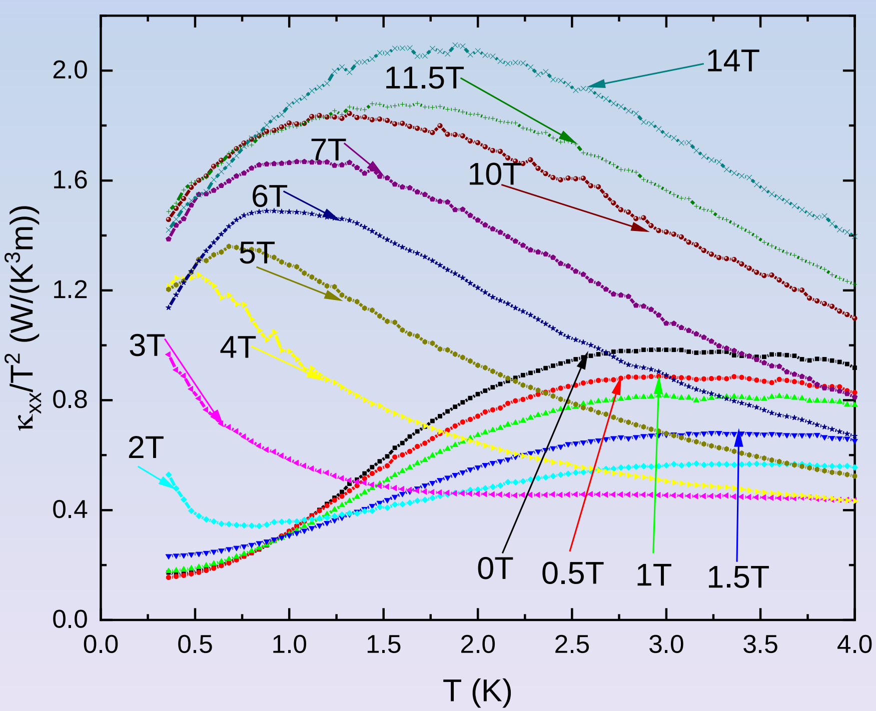


Zhong R., et al. *Under review (2019)*

Thermal conductivity K_{xx}/T



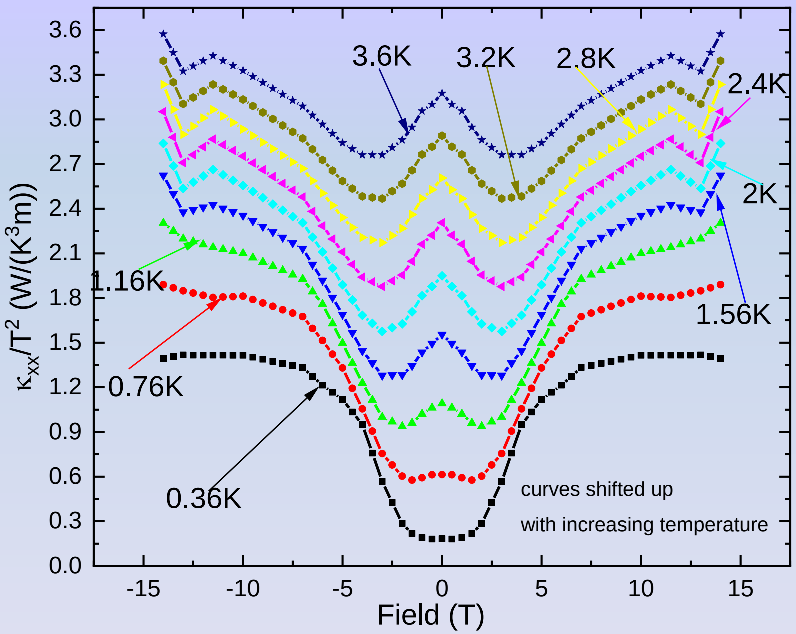
K_{xx}/T^2 **H||c**



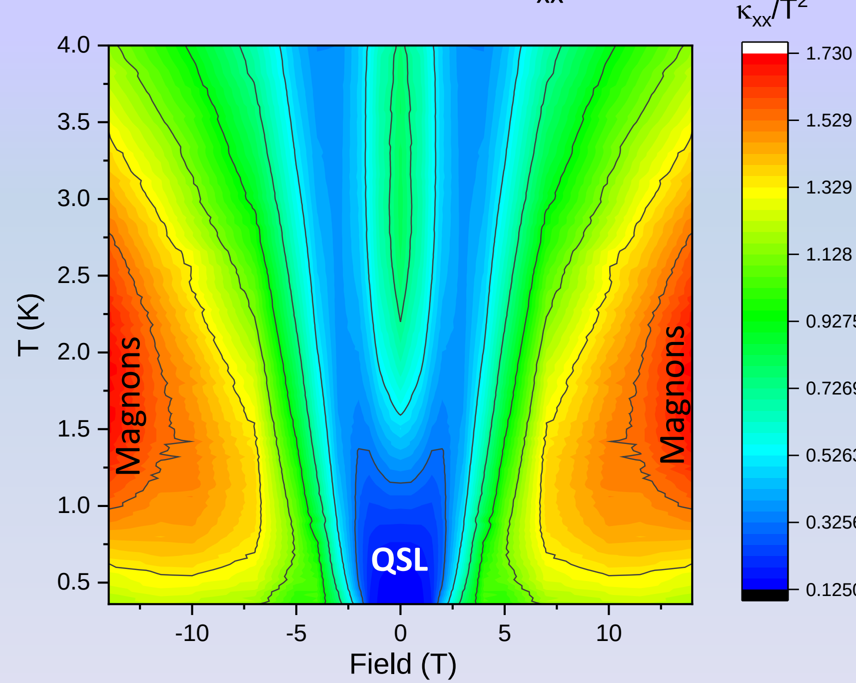
1. Metamagnetic Phase Transition around 2T, consistent with specific heat measurement
2. κ_{xx}/T^2 exhibits soft gap below 2 K

Gao T., et al. *In preparation(2019)*

Thermal conductivity K_{xx}



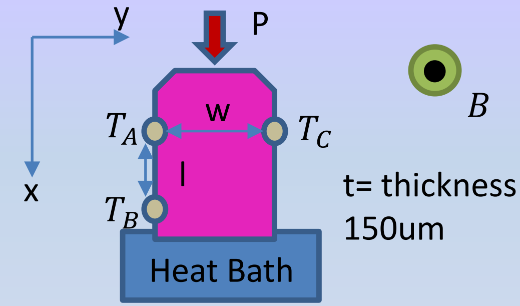
Color (intensity) map of K_{xx}/T^2



Gao T., et al. *In preparation(2019)*

1. Below 2T, heat capacity is very large, but thermal conductivity is small.
2. Above 5 T, the reverse is true.
3. At 0.36K, κ_{xx}^{max} / $\kappa_{xx}^{min}\sim 7.8$, shows huge difference between magnon and QSL(?) regime
4. The spin excitations hold very large entropy, but don't conduct heat (localized?)

Thermal Hall Measurement

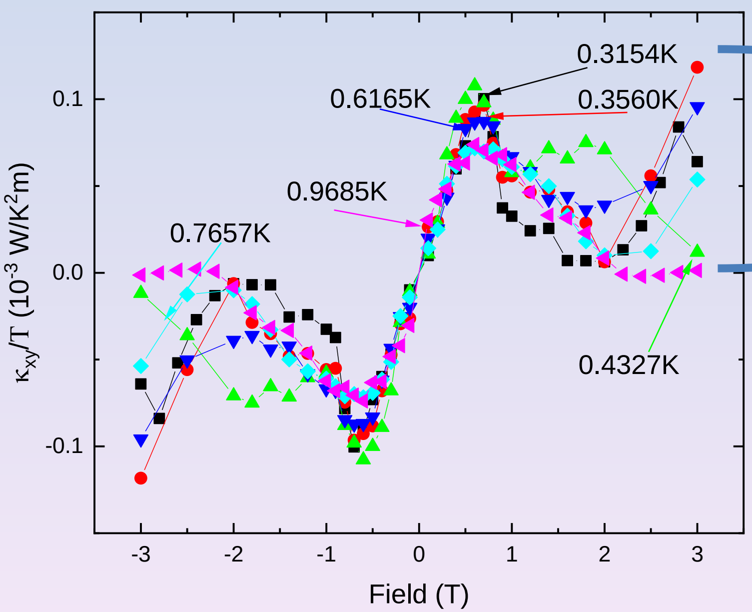


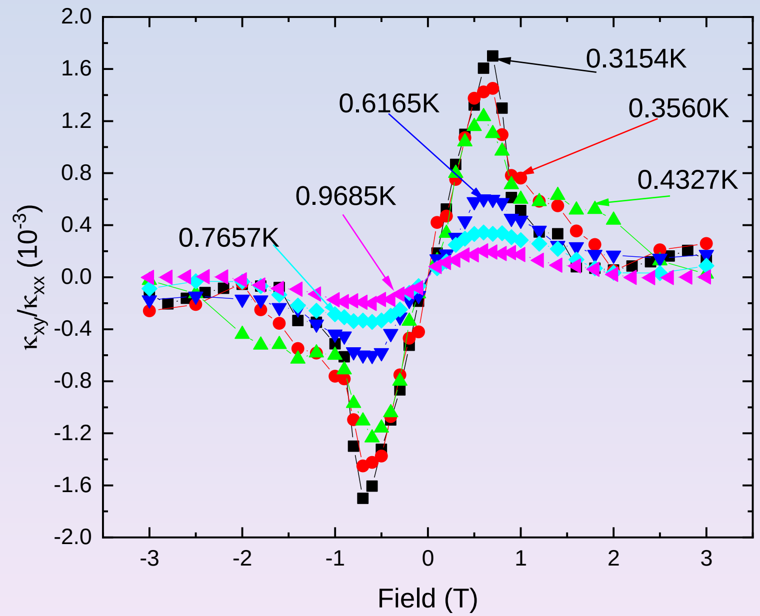
t= thickness
150um

When hall angle is small:

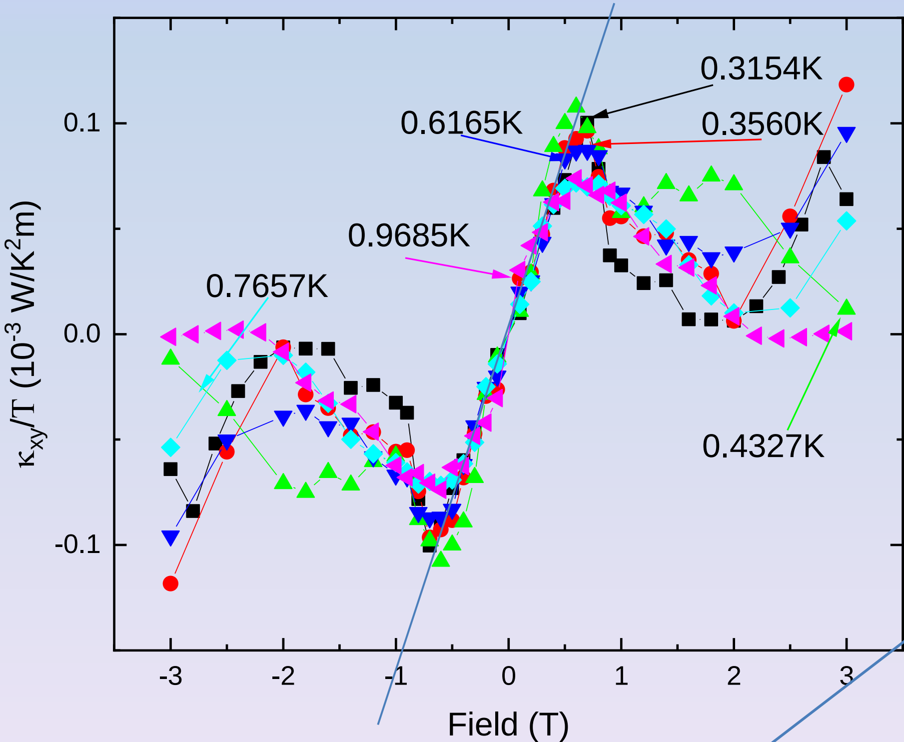
$$\kappa_{xy} \approx \frac{t}{P} \kappa_{xx}^2 \, \Delta T_y$$

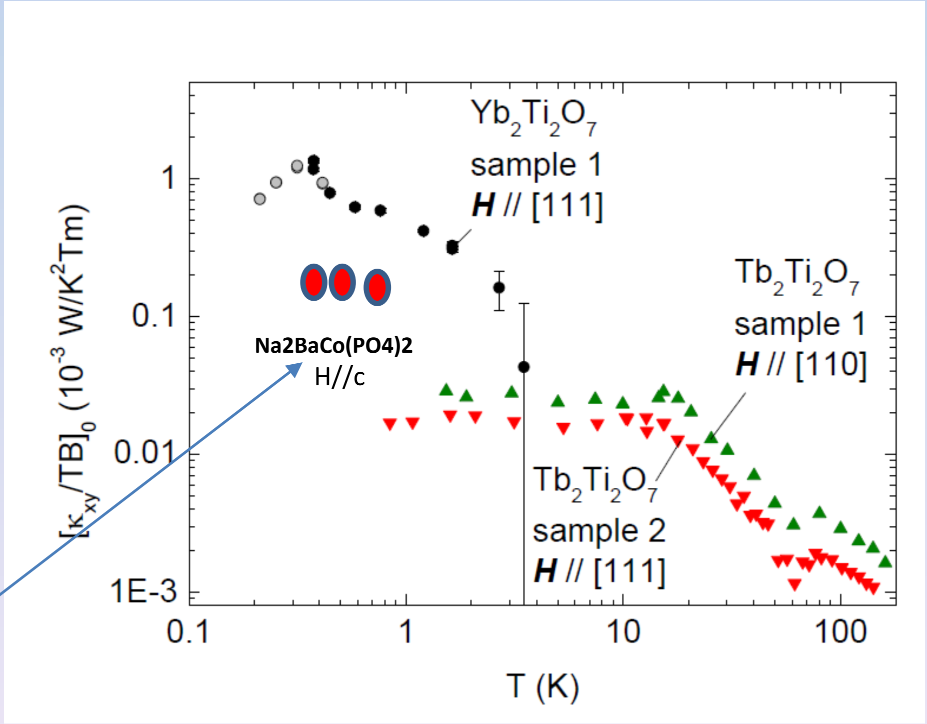
Noise in hall channel increases dramatically with κ_{xx}





Thermal Hall Measurement





κ_{xy}/TB = $1.5\times 10^{-4} W/K^2Tm$

Temperature independent

Summary

In the quantum frustrated state,

1) Spin excitations are increasingly localized as T→0 (poor heat conduction).

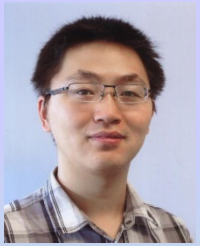
2) A large Kxy appears. From edge states?

3) The spin excitations have massive entropy, but coupling to phonons
   Is strongly suppressed.

4) In $Na_2BaCo(PO_4)_2$, a small gap is observed in Kxx.

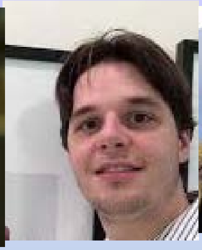
5) A 5-Tesla B field destroys the frustrated state, and all these characteristics.

Max Hirschberger



Tong Gao

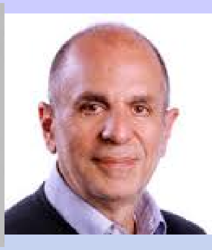
Peter Czajka



Jason Krizan

Ruidan Zhong

Seyed Koohpayeh JHU



Cava

NPO

Observation of a large Hall effect from neutral spin excitations

I) In frustrated magnet $Tb_2Ti_2O_7$

Excitations are not magnons

Are they spinons? Other fractional excitations?

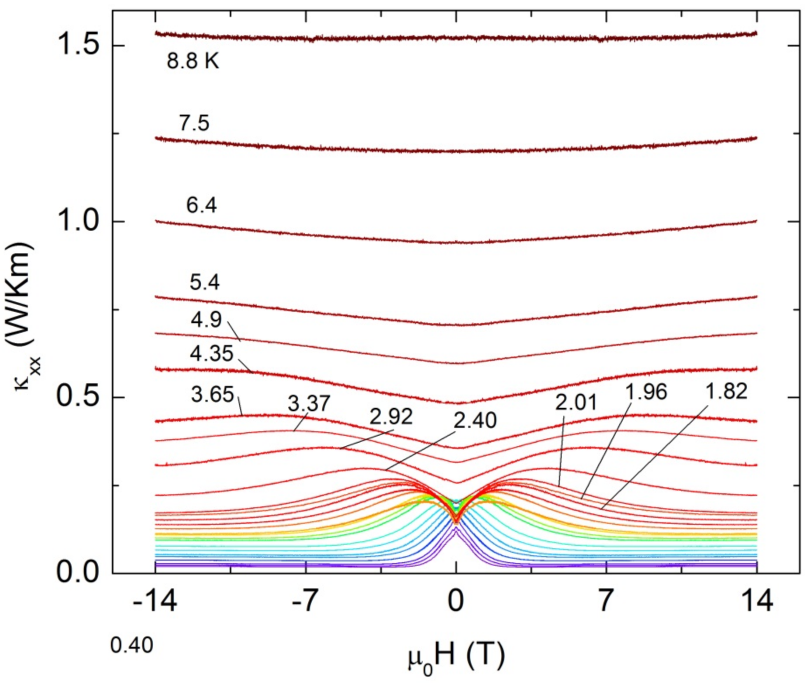
II) In Kagome ferromagnet Cu(1-3, *bdc*)

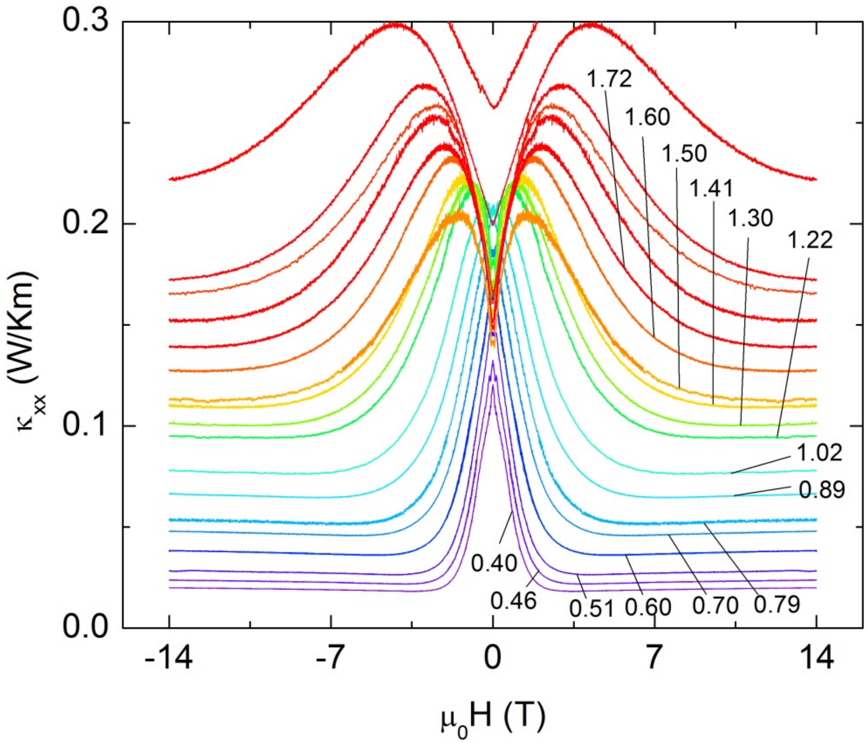
Hall signal observed both above and below Tc

Unexpected sign reversal at Tc

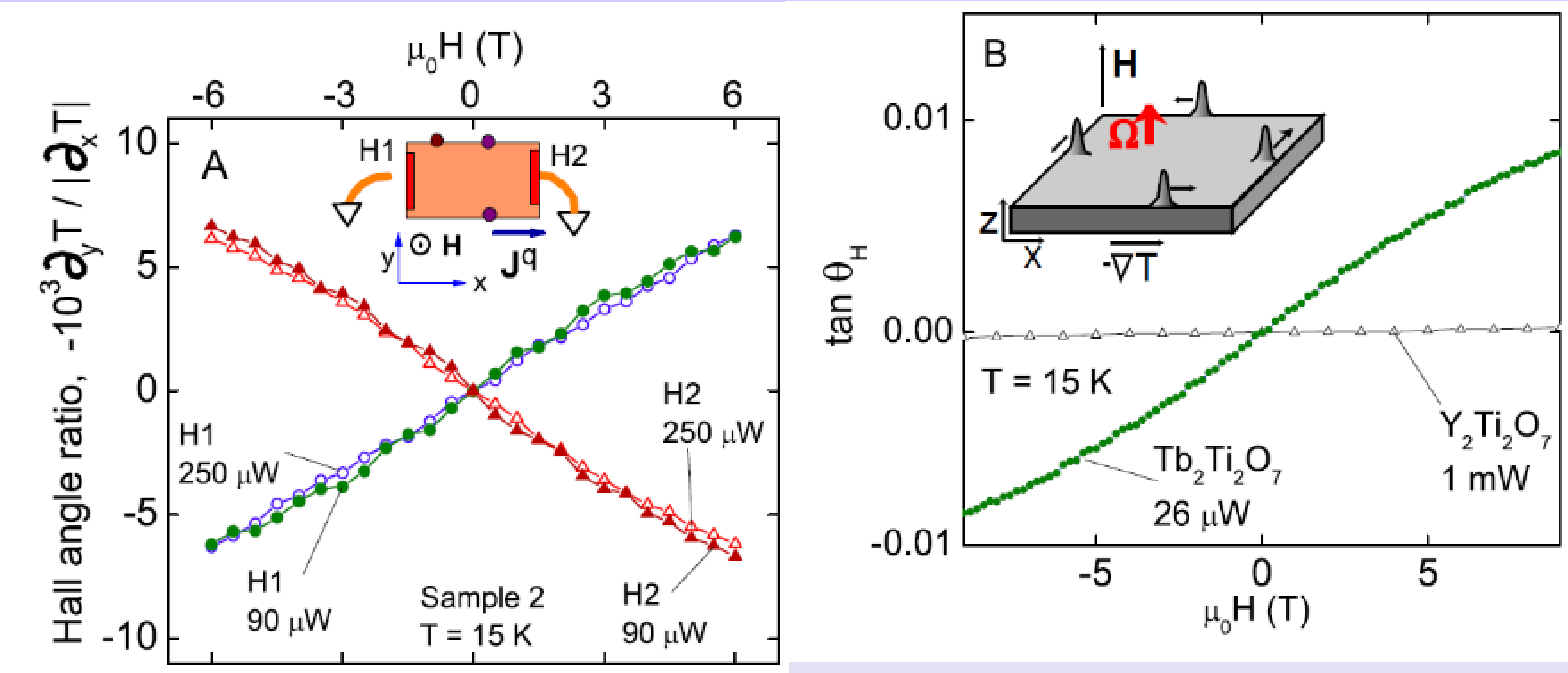
Consequence of Berry curvature in different magnon bands

Experimental checks





Hall effect in a neutral current?   Experimental checks



Checks

Hall signal reverses when gradient (-∇*T*) is reversed in same **B**

Hall signal scales linearly with gradient strength

Hall signal is 1000 x larger than in nonmag analog $Y_2Ti_2O_7$

Observation of the Magnon Hall Effect

Y. Onose,[1,2]* T. Ideue,[1] H. Katsura,[3] Y. Shiomi,[1,4] N. Nagaosa,[1,4] Y. Tokura[1,2,4]

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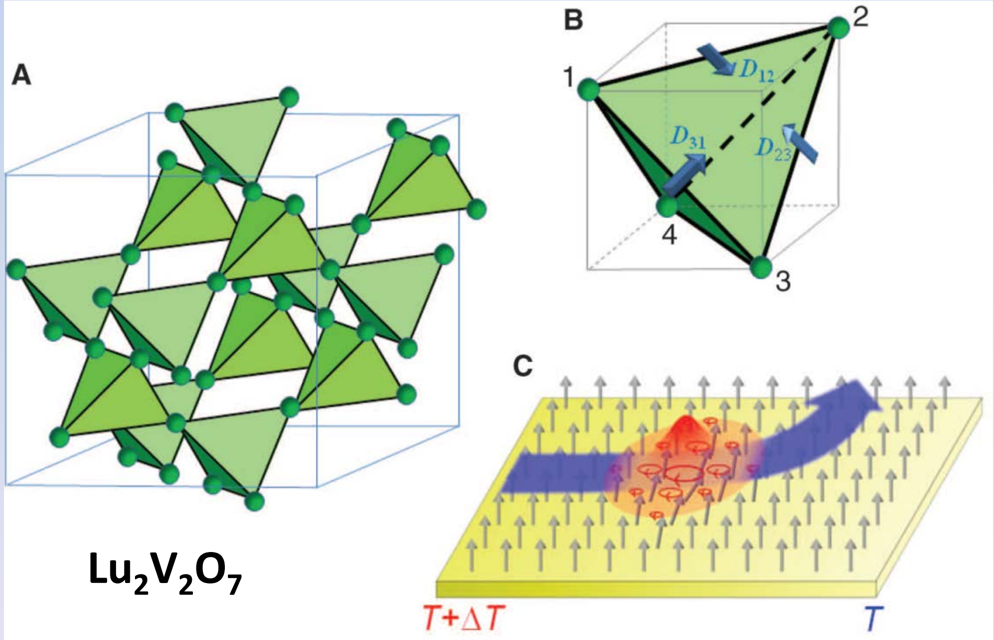
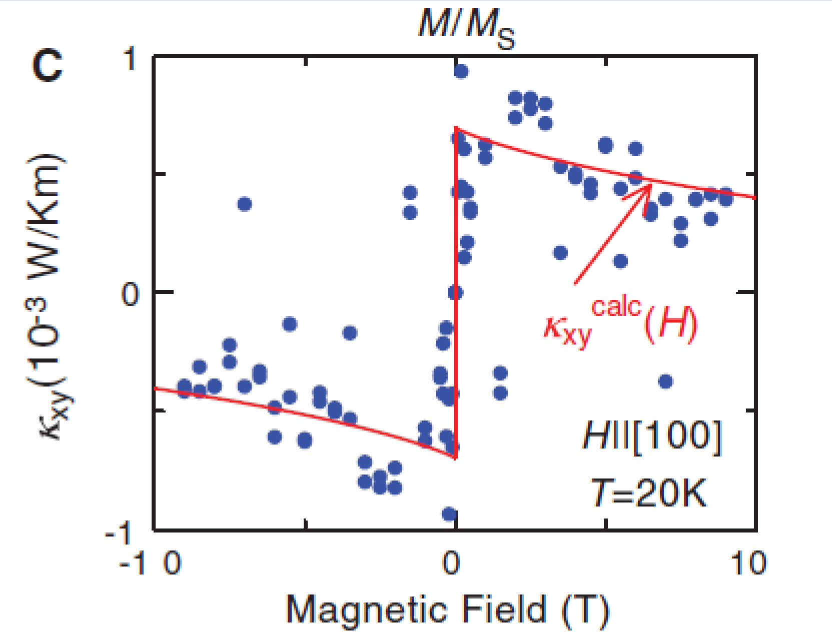
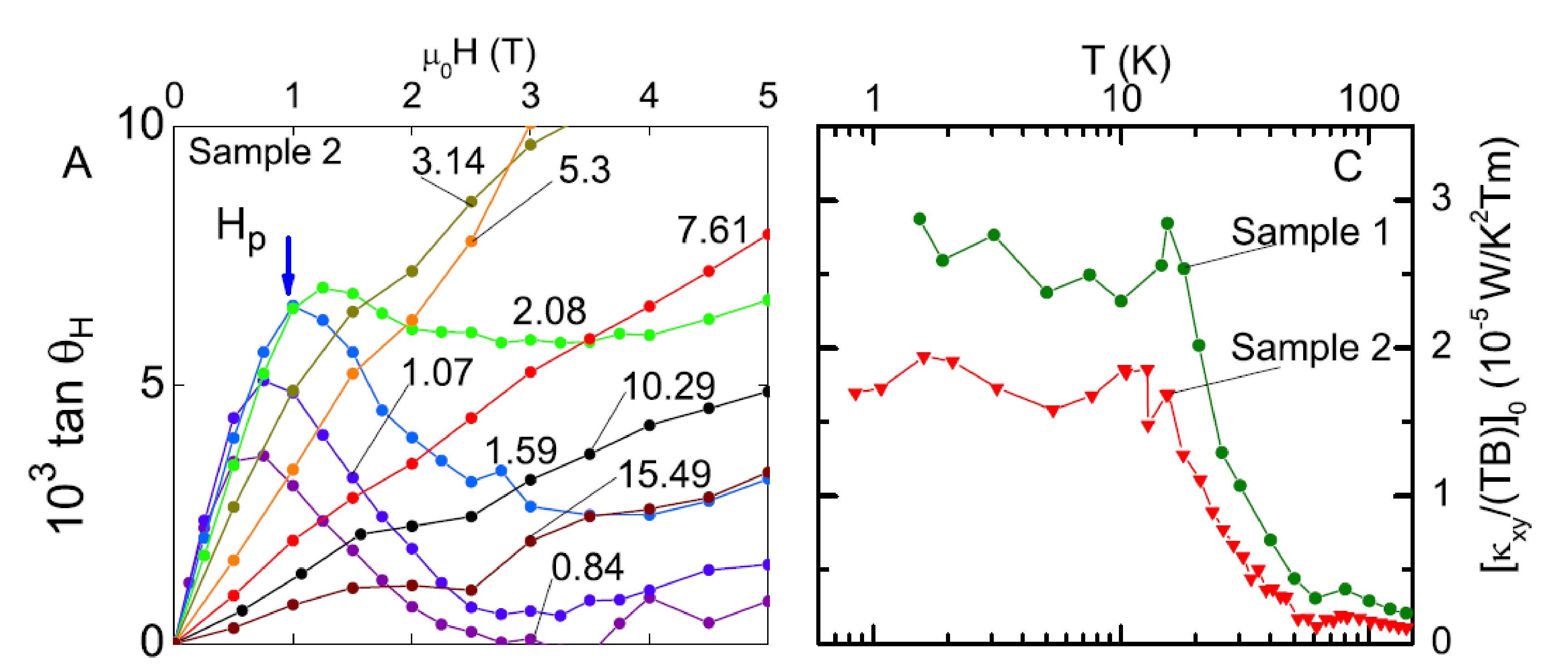




Onose *et al*. observed a weak K_{xy} in insulating pyrochlore ferromagnet $Lu_2V_2O_7$

Katsura, Nagaosa and Lee missed **magnetization** current term (Matsumoto, Murakami)

Hall angle



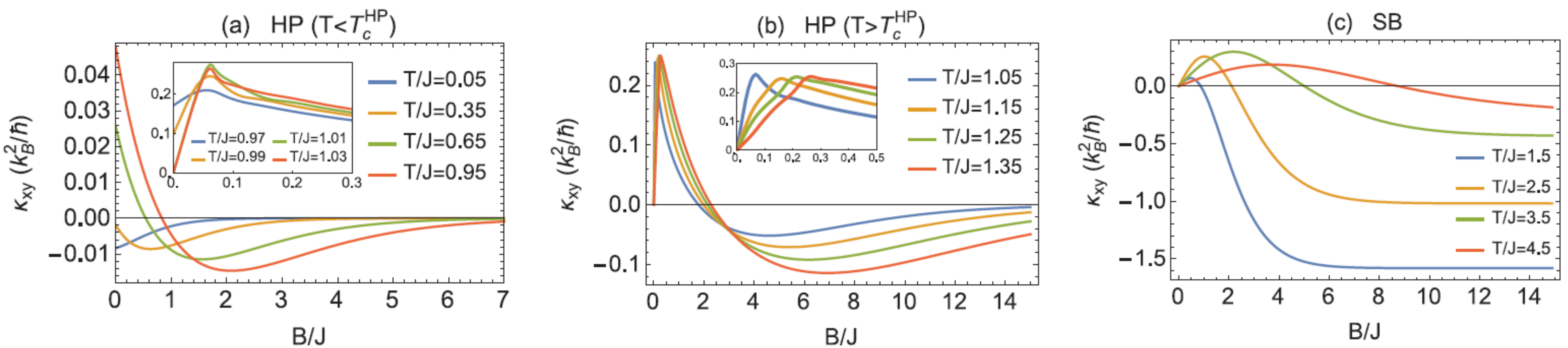
Below 3 K, tan θ_H is H-linear (with constant slope) until $H > H_p$

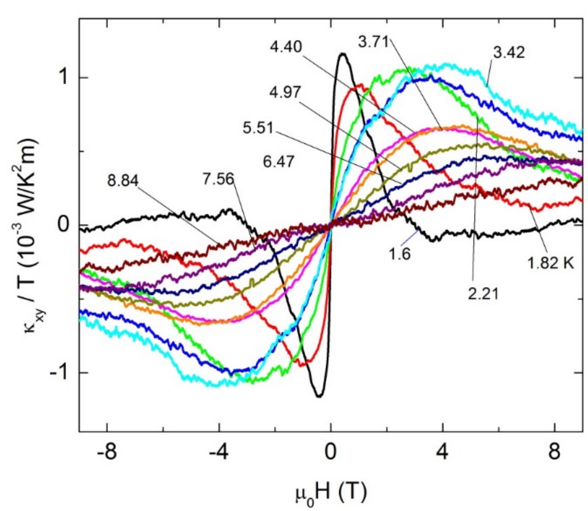
Defines low-*T* state with largest Hall response.

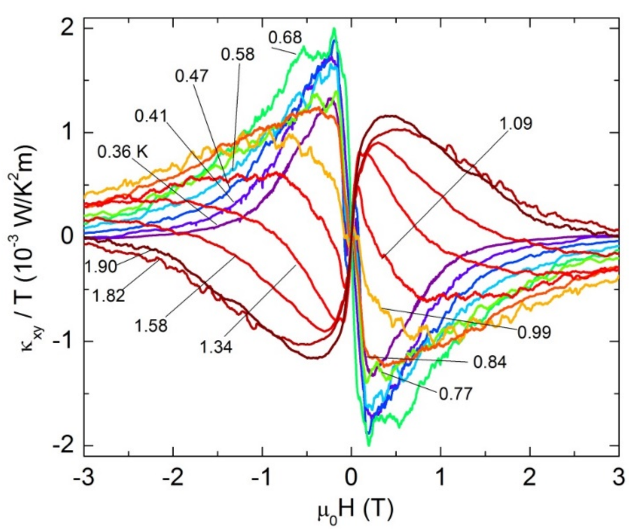
The state is readily destroyed when H exceeds $H_p(T)$

Computed Kxy of Kagome Magnet

Lee, Han and Lee, preprint

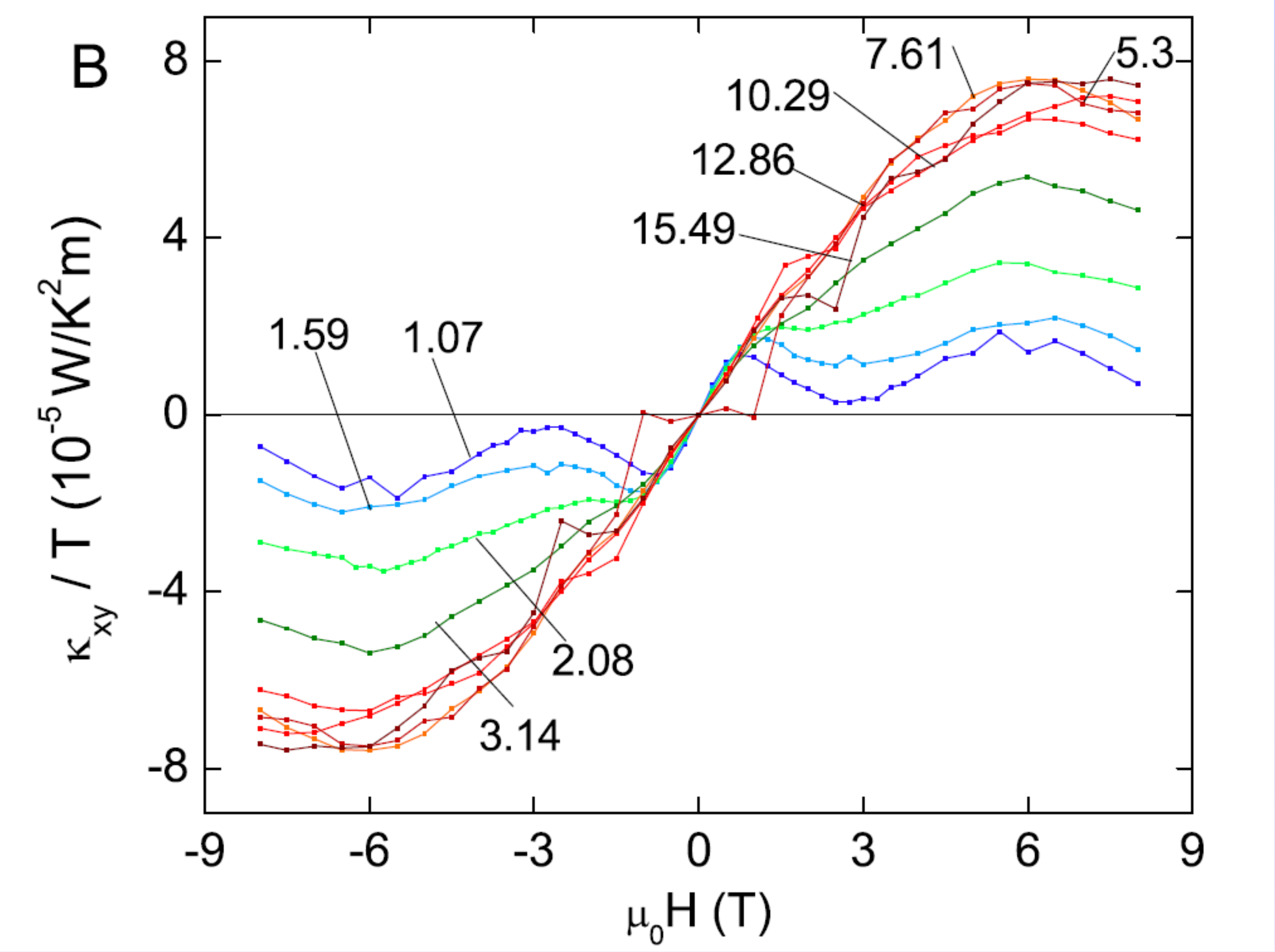






Calculation captures main qualitative features, some quantitative discrepancies

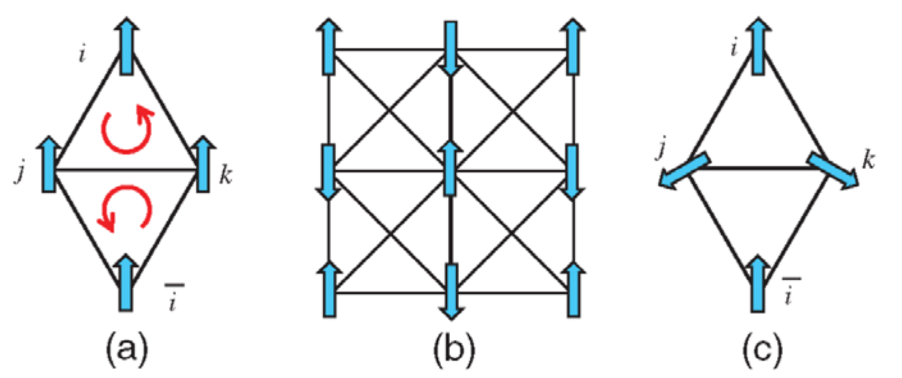
Thermal Hall conductivity 1 < *T* < 15 K

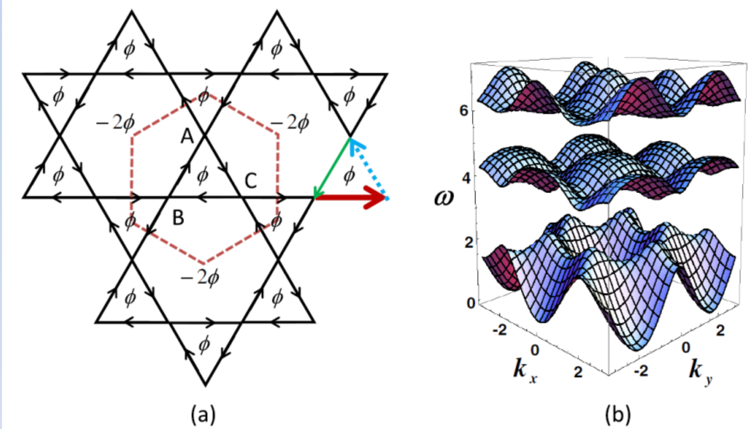


PRL 104, 066403 (2010) PHYSICAL REVIEW LETTERS week ending 12 FEBRUARY 2010

Theory of the Thermal Hall Effect in Quantum Magnets

Hosho Katsura,[1] Naoto Nagaosa,[1,2] and Patrick A. Lee[3]





Pairwise cancellation

Local moments on Kagome lattice

$$H_{\rm ring} = -\frac{24t^3}{U^2}\sin\Phi \vec{S}_i \cdot (\vec{S}_j \times \vec{S}_k),$$

$$\langle \vec{S}_i \rangle \cdot (\delta\vec{S}_j \times \delta\vec{S}_k) + \langle \vec{S}_j \rangle \cdot (\delta\vec{S}_k \times \delta\vec{S}_i) + \langle \vec{S}_k \rangle \cdot (\delta\vec{S}_i \times \delta\vec{S}_j).$$

Predictions:

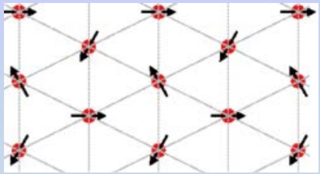
Chiral spin texture leads to K_{xy} in sparse lattices (bonds between asymmetric plaquettes) --- **Kagome** lattice and **pyrochlores**

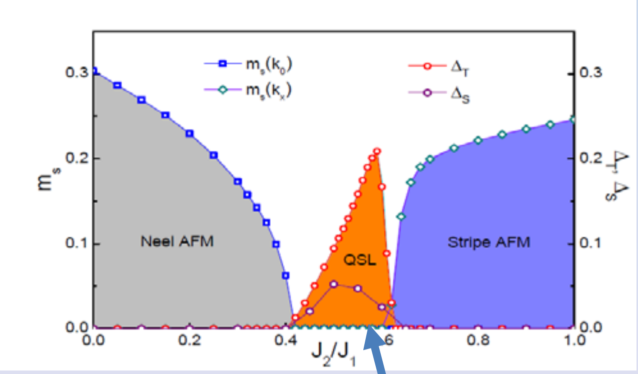
Heuristic derivation of K_{xy} in spin liquid (with fermionic spinons)

Forgot magnetization current?

Triangular Lattice → Heisenberg AFM Nearest Neighbor →

120° Neel State

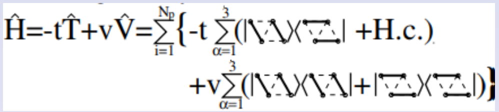




J1- J2 model

$$H = J_1 \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle\langle ij \rangle\rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$

RVB-Quantum Dimmer model



Z2 Topological Quantum Spin Liquid

Ring exchange

$$\hat{H}_{\mathrm{ring}} = J_2 \sum P_{12} + J_4 \sum \left(P_{1234} + P_{1234}^{\dagger} \right)$$

Balents at el, Phys. Rev. B **86**, 024424
Motrunich, Phys. Rev. B **72**, 045105
R. Moessner at el, Phys. Rev. Lett. **86**, 1881

A rough guide to experiments on QSLs

Does it order?

- NMR line splitting
- muSR oscillation
- thermodynamic transition via specific heat, susceptibility
- Bragg peak in neutron/x-ray

Is there a gap?

- Specific heat
- NMR $1/T_1$
- Dynamic susceptibility
- T-dependence of χ

Exotica

- Local measurements
- thermal Hall
- ARPES (on insulator!)
- Proximity effects

Delocalized excitations?

- thermal conductivity
- INS

Structure of excitations?

- E(k) from INS,RIXS
- optics, Raman