Half-integer thermal Hall conductance in a Kitaev spin liquid
– Evidence for chiral Majorana edge current –

Yuichi KASAHARA
Department of Physics, Kyoto University

Half-integer thermal Hall conductance

Chiral Majorana edge current
Collaborators

Takafumi Onishi, Yuji Matsuda
Department of Physics, Kyoto University

Kaori Sugii, Masaaki Shimozawa, Minoru Yamashita
Institute for Solid State Physics, The University of Tokyo

Taka Shibauchi
Department of Advanced Materials Science, The University of Tokyo

Nobuyuki Kurita, Hidekazu Tanaka
Department of Physics, Tokyo Institute of Technology

Joji Nasu
Department of Physics, Tokyo Institute of Technology

Yukitoshi Motome
Department of Applied Physics, The University of Tokyo
1. Introduction: Kitaev quantum spin liquid

2. A candidate of Kitaev magnet $\alpha$-RuCl$_3$

3. Thermal Hall effect in perpendicular fields

4. Thermal Hall effect in tilted fields
   Observation of half-integer thermal Hall conductance

5. Summary

Introduction

Quantum spin liquid (QSL)

Quantum fluctuations melt the long-range magnetic order even at $T = 0$. The ground state with massive entanglement of local spins.

Spin liquid are states which do not break any simple symmetry: Neither spin-rotational symmetry nor lattice translational symmetry.

Platforms of QSL

1D: $S = 1/2$ XXZ chain

2D & 3D: Geometrically frustrated magnets

2D triangular

2D kagome

3D pyrochlore

Exotic physical properties in QSLs

Topological phases
Gauge fluctuations
Fractionalized excitations

Spinon excitation ($S = 1/2, e = 0$)
Kitaev model

\( S = 1/2 \) spins on tri-coordinate lattices

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

Kitaev Interaction

Bond-dependent Ising-like interaction

Exchange frustration

Honeycomb lattice (2D)


Hyper-honeycomb lattice (3D)

\( \mathcal{H}_K = -J_x \sum_{<ij>_x} S_i^x S_j^x - J_y \sum_{<ij>_y} S_i^y S_j^y - J_z \sum_{<ij>_z} S_i^z S_j^z \)

Jordan-Wigner transformation

Majorana representation

\( W_p = -1 \)

Free Majorana fermions on a honeycomb lattice


Fractionalization of quantum spins

Itinerant Majorana fermion \( c_i \)

Localized Majorana fermion \( \bar{c}_i \)

\( W_p = \eta_r \eta_{r'} \)

Spin fractionalization occurs below \( \sim J_K/k_B \).

(proximate spin liquid state)

Two types of QSLs

Gapped QSL: Toric code

Gapless QSL: Majorana metal
Spin-orbit assisted Mott insulator with $j = 1/2$

$4d^5$ or $5d^5$

d-orbitals

<table>
<thead>
<tr>
<th>Octahedral crystal field</th>
<th>$\epsilon_g$</th>
<th>$\sim 3\text{eV}$</th>
<th>spin-orbit coupling</th>
<th>$\vec{L} \cdot \vec{\sigma}$</th>
<th>$j = 1/2$, $\sim 400\text{meV}$</th>
<th>$j = 3/2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>IrO$_6$ cage</td>
<td>$t_{2g}$</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>$U$</td>
</tr>
</tbody>
</table>

90° bond formed by edge-shared octahedra

Candidate materials

2D honeycomb lattice

Na$_2$IrO$_3$

Y. Singh & P. Gegenwart, PRB 82, 064412 (2010).

3D hyper-honeycomb lattice

β-Li$_2$IrO$_3$


α-RuCl$_3$

K. W. Plumb et al., PRB 90, 041112 (2014).
Layered honeycomb magnet $\alpha$-RuCl$_3$

$$\mathcal{H} = \sum_{\langle ij \rangle} \left[ J \vec{S}_i \cdot \vec{S}_j + J_K S_i^\gamma S_j^\gamma + \Gamma (S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha) \right]$$

- Heisenberg
- Kitaev
- off-diagonal exchange

$$J = -1.7 \text{ meV}$$
$$K = -6.7 \text{ meV}$$
$$\Gamma = +6.6 \text{ meV}$$

Dominant Kitaev term $J_K/k_B \sim 100 \text{ K}$

$$K_x = -6.7 \text{ meV}, K_y = -6.7 \text{ meV}, K_z = -5.0 \text{ meV}$$

S. M. Winter et al., PRB 93, 214431 (2016).

Presence of non-Kitaev interaction

- AFM order with zigzag spin structure at $T_N \sim 7.5 \text{ K}$
- Transition at 14 K appears due to stacking faults.
Possible signatures of Kitaev QSL in $\alpha$-RuCl$_3$

**Raman scattering**

![Raman scattering plot](image)


**Inelastic neutron scattering**

![Inelastic neutron scattering plots](image)

S.-H. Do *et al.*, Nat. Phys. [10.1038/nphys4298](http://doi.org/10.1038/nphys4298).

**Broad magnetic continuum** appears below $\sim J_K/k_B$

**Possible signature of spin fractionalization**

More direct measurements are required.

Broad magnetic continuum at high energy

Fermionic excitations

What gives direct signature of Majorana fermions?

Effect of magnetic field \((h || [111])\) 

\[ \mathcal{H} = \mathcal{H}_K + \mathcal{H}_h^{\text{eff}} \]

\[ \mathcal{H}_K = -J_x \sum_{<ij>_x} S_i^x S_j^x - J_y \sum_{<ij>_y} S_i^y S_j^y - J_z \sum_{<ij>_z} S_i^z S_j^z \]

\[ \mathcal{H}_h^{\text{eff}} = -\tilde{\hbar} \sum_{(ijk)} S_i^x S_j^y S_k^z \]

\[ \tilde{\hbar} = \lambda h^3 \sim \frac{h^3}{\Delta_f^2} \]

Flux gap \(\Delta_f \sim 0.06 J_K\)

Topological system characterized by Chern insulator under \(H\)

\[ \text{Chiral edge current of Majorana fermions} \]
What gives direct signature of Majorana fermions?

**Integer QHE**

Chiral edge current of electrons

\[ \sigma^{2D}_{xy} = \nu \frac{e^2}{\hbar} \quad \nu : \text{Chern number} \]
\[ = \# \text{ of chiral edge modes} \]
\[ \kappa^{2D}_{xy} = \nu \frac{\pi k_B^2}{6 \hbar} \]

**Kitaev QSL**

Chiral edge current of charge neutral *Majorana fermions*

\[ \frac{\kappa^{2D}_{xy}}{T} = q \frac{\pi k_B^2}{6 \hbar} \quad q : \text{Central charge} \]
\[ q = \nu \text{ in IQHE} \]

\[ q = \frac{\nu}{2} \quad \frac{\kappa^{2D}_{xy}}{T} = \frac{1}{2} \left( \frac{\pi k_B^2}{6 \hbar} \right) \]

Half-integer thermal Hall conductance in a Kitaev QSL
Thermal Hall effect in insulating magnets

\[
\begin{pmatrix}
q \\
0
\end{pmatrix} =
\begin{pmatrix}
\kappa_{xx} & \kappa_{xy} \\
-\kappa_{xy} & \kappa_{yy}
\end{pmatrix}
\begin{pmatrix}
-\nabla T_x \\
-\nabla T_y
\end{pmatrix}
\]

Ferromagnetically ordered state

Lu$_2$V$_2$O$_7$
Ho$_2$V$_2$O$_7$, In$_2$Mn$_2$O$_7$, BiMnO$_3$
Cu(1-3,bdc)

Ideue et al., PRB 85, 134411 (2012).

Paramagnetic state

Tb$_2$Ti$_2$O$_7$


Spin liquid state

Cu$_3$V$_2$O$_7$(OH)$_2$·2H$_2$O

D. Watanabe, PNAS 113, 8653 (2016).

Magnon Hall effect arising from Berry phase

\[
\kappa_{xy} = \frac{k_B T}{\hbar V} \sum_k \sum_n \left\{ c_2[g(\varepsilon_{nk})] - \frac{\pi^2}{3} \right\} \Omega_{nk}
\]

Spinon Hall effect in QSL state with spinon Fermi surface

\[
\begin{align*}
\kappa_{\text{spinon}}^{xx} &= 2 \frac{\pi^2}{3} \left( \frac{\varepsilon_F}{\hbar \tau} \right) \frac{k_B^2 T}{\hbar} \frac{1}{d} \\
\kappa_{\text{spinon}}^{xy} &= \kappa_{xx}^{\text{spinon}} \left( \omega_c \tau \right)
\end{align*}
\]

H. Katsura et al., PRL 104, 066403 (2010).
Thermal transport measurements in $\alpha$-RuCl$_3$

Thermal Hall effect

\[
\begin{pmatrix}
q \\
0
\end{pmatrix} =
\begin{pmatrix}
\kappa_{xx} & \kappa_{xy} \\
-\kappa_{xy} & \kappa_{xy}
\end{pmatrix}
\begin{pmatrix}
-\nabla T_x \\
-\nabla T_y
\end{pmatrix}
\]

- **Spin liquid state:**
  Kagome volborthite Cu$_3$V$_2$O$_7$(OH)$_2$·2H$_2$O
  \( \kappa_{xy}/T \sim 10^{-5} \text{ W/K}^2\text{m} \) spinon

- **Magnetically ordered state:**
  Kagome Cu-(1-3,bdc) \( \kappa_{xy}/T \sim 10^{-5} - 10^{-4} \text{ W/K}^2\text{m} \) magnon
  Pyrochlore Lu$_2$V$_2$O$_7$

**α-RuCl$_3$ single crystals**

- Magnetic susceptibility
- Specific heat

- Clear anomaly at \( T_N \sim 7.5 \text{ K} \)
- No discernible anomaly at \( \sim 14 \text{ K} \) due to stacking faults

High quality single crystal
Longitudinal thermal conductivity $\kappa_{xx}$

- Clear anomaly in $\kappa_{xx}$ at $T_N$
- Suppression of $\kappa_{xx}$ by magnetic field $\leftrightarrow \kappa_{xx}^{ph}$ is usually enhanced due to suppression of spin-phonon scattering by spin polarization.

Thermal transport is governed by *spin excitations*.

However, it is difficult to separate spin & phonon contributions.

Thermal Hall effect
Thermal Hall conductivity $\kappa_{xy}$

Finite $\kappa_{xy} \sim 10^{-2}$ W/Km at $T < J_K/k_B$

e.g.) $\kappa_{xy} < 10^{-3}$ W/Km

in volborthite (spin liquid)
Tb$_2$Ti$_2$O$_7$ (paramagnet)

Distinct $H$-dependence below and above $T_N$

- Sign change below $T_N$
- Upward curvature above $T_N$
but downward below $T_N$

Thermal Hall effect below and above $T_N$ is different in origin.
Thermal Hall conductivity $\kappa_{xy}$

- **Phonons**
  $\kappa_{xy}/T \sim 10^{-6}$ W/K$^2$m
  Different $T$-dependence

- **Magnons**
  Finite $\kappa_{xy}/T$ usually appears in the ordered state.
  Small DM interaction
  $D/k_B \sim 5$ K $\ll J/k_B \sim 80$ K
  S. M. Winter et al., PRB 93, 214431 (2016).

- **Spin liquid with spinon Fermi surface**
  In volborthite, Hall signal is negative.
  $\kappa_{xy}/T \sim 10^{-5}$ W/K$^2$m

- **Enhancement of $\kappa_{xy}$ with positive sign** below $J_K/k_B \sim 80$ K
- **Broad peak at $\sim 20$ K**

- **Exotic quasiparticle excitations inherent to the spin-liquid state of $\alpha$-RuCl$_3$.**
Comparison with numerical calculations

$T$-dependence is consistent with numerical calculations for the 2D pure Kitaev model.

- Enhancement of $\kappa_{xy}$ with positive sign below $T < J_K/k_B$
- Broad peak at $T \sim 0.1J_K/k_B$
- $\kappa_{xy}/T$ reaches close to half of the quantization value.

Possible signature of Majorana fermion excitations

Comparison with numerical calculations

\[ \kappa_{xy}/T \text{ (10}^{-4}\text{ W/K}^2\text{m)} } \]

\( T \)-dependence is consistent with numerical calculations for the 2D pure Kitaev model.

- Enhancement of \( \kappa_{xy} \) with \textbf{positive sign} below \( T < J_K/k_B \)
- Broad peak at \( T \sim 0.1J_K/k_B \)
- \( \kappa_{xy}/T \) reaches close to \textit{half of the quantization value}.

\textbf{Possible signature of Majorana fermion excitations}

However, quantization of \( \kappa_{xy}^{2D}/T \) is not attained due to the magnetic order.

Suppression of AFM order by in-plane fields

Low-temperature properties are masked by the magnetic order.

Key questions:
Is the magnetic order suppressed by tuning parameters?
Whether Kitaev QSL survives when suppressing the order?

AFM order is little influenced by out-of-plane fields, but it is easily suppressed by in-plane fields.

\[ H_{c||} \sim 7-8 \, \text{T} \]

M. Majumder et al., PRB 91, 180401(R) (2015).
Suppression of AFM order by in-plane fields

Low-temperature properties are masked by the magnetic order.

Key questions: Is the magnetic order suppressed by tuning parameters? Whether Kitaev QSL survives when suppressing the order?

\[ T^1_s \propto \frac{1}{T} \exp \left(-\frac{n\Delta}{T}\right) \]

NMR: Unusual spin gap


NMR:}

\[ H_{c||} \sim 7-8 \, \text{T} \]
Suppression of AFM order by in-plane fields

Low-temperature properties are masked by the magnetic order.

Key question: Kitaev QSL survives when suppressing the magnetic order?

**Kitaev QSL emerges under in-plane magnetic fields.**

Neutron scattering: Magnetic continuum at high energy above $H_{c||}$
What gives direct signature of Majorana fermions?

Low-temperature properties are masked by the magnetic order.

**Key questions:**
- The magnetic order can be suppressed by tuning parameters?
- Kitaev QSL survives when suppressing the magnetic order?

**Thermal Hall effect in a Kitaev QSL state**

**Measurements of thermal Hall effect in tilted fields**

Phase transition is tuned by \( H_{||} = H\sin \theta \).
Thermal Hall response is determined by \( H_{\perp} = H\cos \theta \).
Longitudinal thermal conductivity $\kappa_{xx}$

Strongly anisotropic response: Quasi 2D nature of magnetic properties.

Suppression of the AFM order by in-plane field component.
Measurements of thermal Hall effect in a Kitaev magnet candidate $\alpha$-RuCl$_3$

**Perpendicular fields**

Striking enhancement of $\kappa_{xy}/T$ with positive sign below $T \sim J_K/k_B$

A broad peak at $T \sim 0.1J_K/k_B$

**Signature of Majorana fermion excitations**


**Tilted fields**

Observation of *half-integer thermal Hall conductance* for the first time.

**Evidence for chiral Majorana edge current**

Sudden disappearance of the quantum Hall plateau at high field

**Topological phase transition**