Electrical manipulations of Kitaev spin liquids

Reinhold Egger

Institut für Theoretische Physik

Heinrich Heine Universität Düsseldorf

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Overview

- **Introduction**
  - Kitaev honeycomb model in magnetic field: Non-Abelian chiral spin liquid
  - Ising anyon: Z_2 vortex with Majorana zero mode (MZM)
  - Kitaev materials, e.g., α-$RuCl_3$
  - Electric polarization effects in Mott insulators

- **Local charge imbalance in Kitaev materials near Z_2 vortices**
  Charge density profile around vortex is related to spin correlations
  → generates quadrupolar electric field

- **Trapping & moving vortices by STM/AFM tips**

- **Energy absorption spectroscopy**
  ⟶ toward detection of MZMs in Kitaev spin liquids

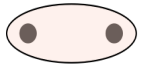
R. G. Pereira and R. Egger, PRL (in press)
arXiv:2008.05334

Quantum spin liquids

Savary & Balents, Rep. Prog. Phys. 2017
Zhou, Kanoda & Ng, Rev. Mod. Phys. 2019

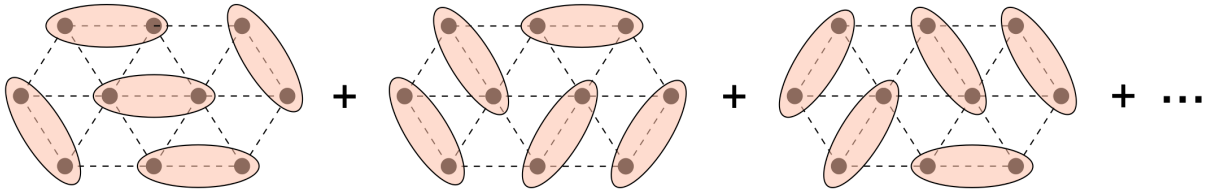
Exotic long-range entangled phase of quantum magnets (here: spin S=½) with frustrated interactions

Ground state: massive superposition of resonating valence bond (RVB) type solids formed by spin singlets



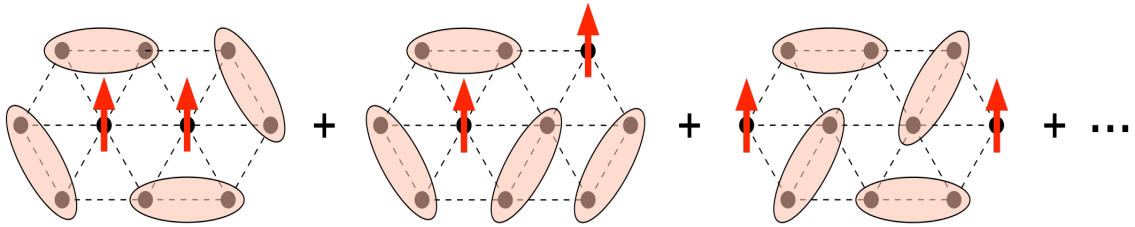
$$= \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)$$

$$|\Psi_0\rangle =$$



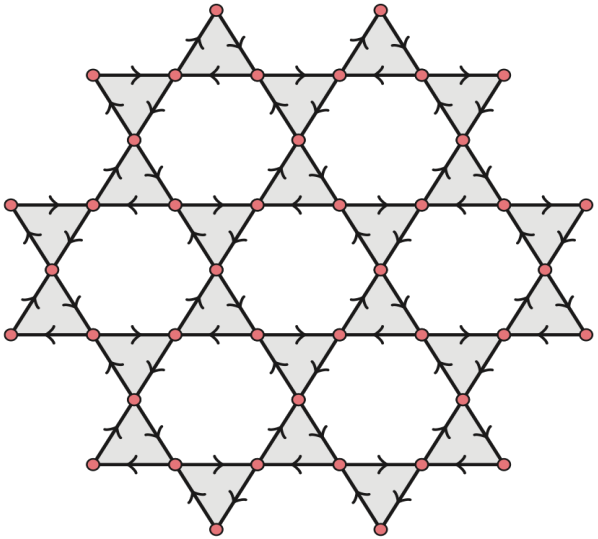
→ fractionalized excitations (e.g. spinons, anyons,…)

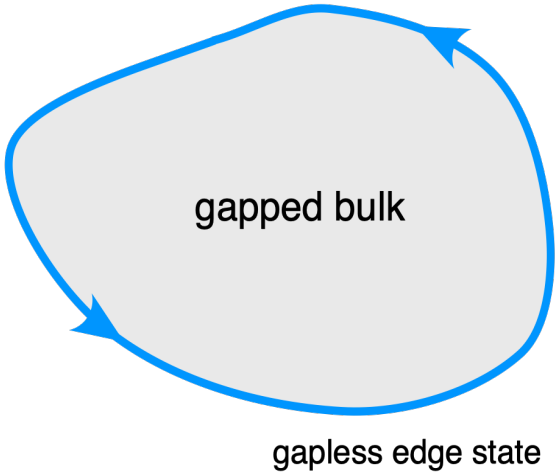
$$|\Psi_{\text{exc}}\rangle =$$



Chiral spin liquids

Example: **Kalmeyer-Laughlin state** (equivalent to bosonic QH fluid at $\nu = ½$) for 2D Kagome lattice with chiral three-spin interactions → spinons with semionic statistics & chiral gapless edge boson mode





$$H = J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_\chi \sum_{i,j,k \in \triangle} \mathbf{S}_i \cdot (\mathbf{S}_j \times \mathbf{S}_k)$$

Bauer et al., Nat. Comm. 2016;
Ferraz, Ramos, RE & Pereira, PRL 2019

Kitaev honeycomb model

Kitaev, Ann. Phys. 2006

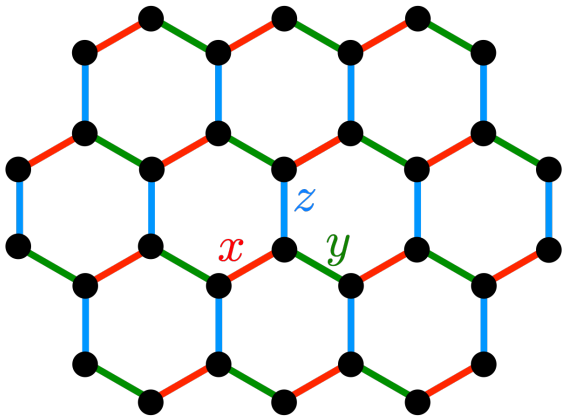
$$H = -K \sum_{\langle jl \rangle_\alpha} S_j^\alpha S_l^\alpha - \kappa \sum_{\langle jk \rangle_\alpha, \langle kl \rangle_\beta} S_j^\alpha S_k^\gamma S_l^\beta$$

$(\alpha\beta\gamma)$: cyclic permutation of *(xyz)*
(jk),(kl): adjacent n.n. bonds

Kitaev exchange coupling
between n.n. sites (j,l)
bond type : $\alpha = x, y, z$
Here: symmetric FM case, K>0

due to magnetic field,
preserves integrability!

→ **spin exchange frustration**



Kitaev model exactly solvable via Majorana representation of spins

For κ>0: gapped non-Abelian chiral spin liquid
→ Ising anyon (Z_2 vortex with MZM) & chiral Majorana edge mode

Majorana representation

Kitaev, Ann. Phys. 2006

For each site j, use four Majorana operators $c_j, c_j^{\alpha=x,y,z}$ with local parity constraint $D_j \equiv c_j c_j^x c_j^y c_j^z = 1$

→ Spin-1/2 operators are written as $S_j^\alpha = \frac{i}{2} c_j c_j^\alpha$

For every n.n. bond: conserved bond operator

= static Z_2 gauge field $u_{\langle jl \rangle_\alpha} \equiv i c_j^\alpha c_l^\alpha = -u_{\langle lj \rangle_\alpha} = \pm 1$

For given gauge field configuration $|G\rangle$: Kitaev model → noninteracting Majorana Hamiltonian for $\{c_j\}$

$$H_{|G\rangle} = \frac{iK}{4} \sum_{\langle jl \rangle_\alpha} u_{\langle jl \rangle_\alpha} c_j c_l - \frac{i\kappa}{8} \sum_{\langle jk \rangle_\alpha, \langle kl \rangle_\beta} u_{\langle jk \rangle_\alpha} u_{\langle kl \rangle_\beta} c_j c_l$$

Diagonalization → eigenstates $|M(G)\rangle$

Kitaev model: Eigenstates

Projection to ensure local parity constraint → eigenstates:

$$\left|\Psi\right\rangle = \hat{\mathrm{P}}\left|M(G)\right\rangle \otimes \left|G\right\rangle \qquad \hat{\mathrm{P}} = \prod_j \frac{1+D_j}{2}$$

Spin op's (and H) invariant under Z_2 gauge transformations

$$\left(c_j, c_j^\alpha\right) \mapsto \left(-c_j, -c_j^\alpha\right)$$

Gauge-invariant Z_2 flux through hexagon #*p:*

$$W_p = 2^6 S_1^z S_2^x S_3^y S_4^z S_5^x S_6^y = \prod_{\langle jl\rangle_\alpha \in p} u_{\langle jl\rangle_\alpha} = \pm 1$$

Ground state: $\forall p: W_p = 1 \quad \rightarrow$ choose gauge $u_{\langle jl\rangle_\alpha} = +1$

$\rightarrow$ H transl. invariant, solve by Fourier transform.

Z_2 vortex (vison) = plaquette with $W_p = -1$

Excitations of non-Abelian Kitaev spin liquid

Kitaev, Ann. Phys. 2006

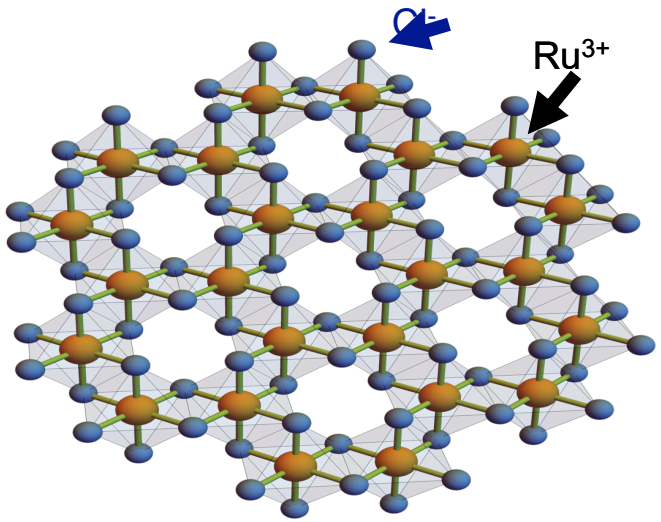
- Vison harbors a MZM: Ising anyon
- Bulk excitations:
  - gapped neutral Majorana fermions
  - Ising anyons
- Chiral Majorana edge mode at boundary
- Close analogy to 2D chiral p-wave superconductors!
- MZMs trapped at Z_2 vortices could (eventually) be useful for topological quantum information processing

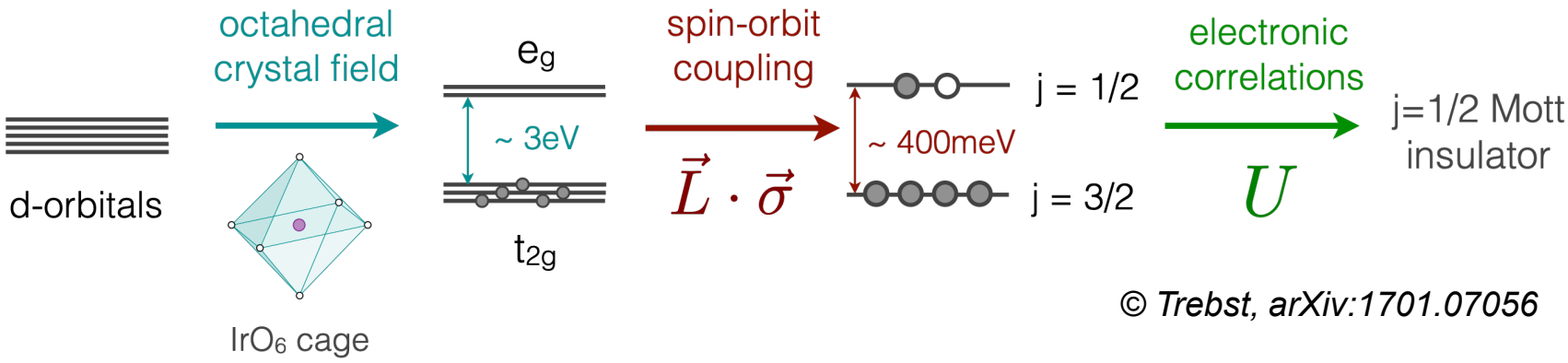
Kitaev materials

Jackeli & Khaliullin, PRL 2009
Trebst, arXiv:1701.07056
Winter et al., J. Phys. Cond. Matt. 2017

Dominant Kitaev interactions can be generated in spin-orbit assisted Mott insulators where strong spin-orbit coupling causes effective angular momentum j=1/2

Examples: Iridates, α-$RuCl_3$, …



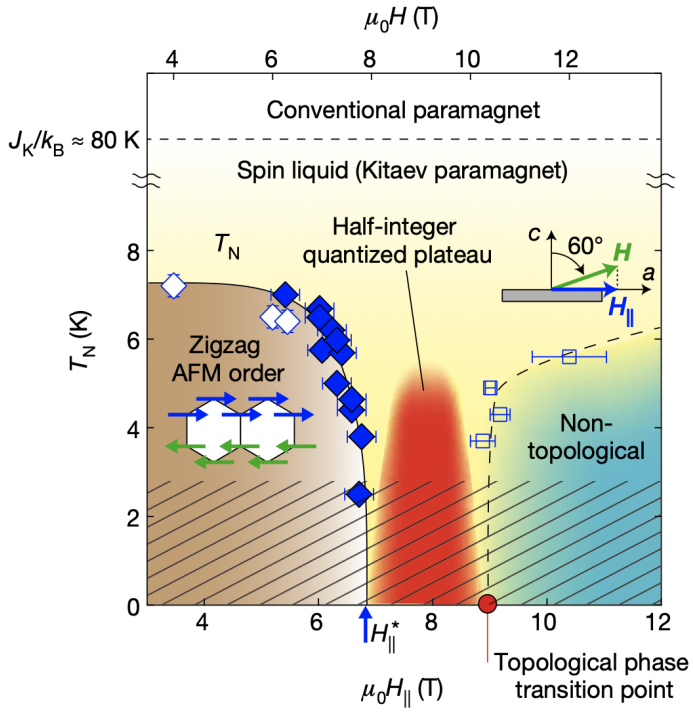


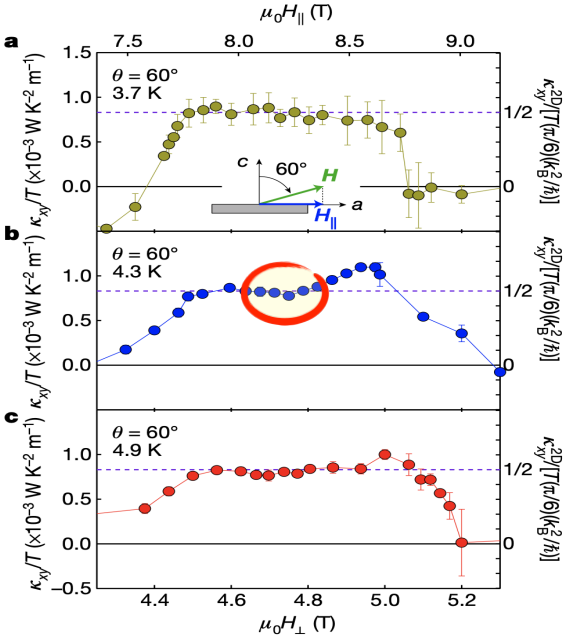
© *Trebst, arXiv:1701.07056*

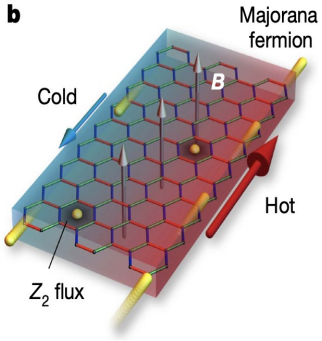
Evidence for Kitaev spin liquid

Quantized thermal Hall conductivity as experimental evidence for chiral Majorana edge mode in α-$RuCl_3$ at intermediate magnetic fields

Kasahara et al., Nature 2018







existence of spin liquid phase still debated

Other evidence: structure factor, Raman, …

Electric detection methods?

Idea: Mott insulator eigenstates may harbor nontrivial electric polarization profile (in a charge insulator!)

Bulaevski, Batista, Mostovoy, and Khomskii, PRB 2008
Khomskii, J. Phys. Cond. Matt. 2010

Local charge imbalance operator can be expressed in terms of the low-energy spin degrees of freedom

→ spin-spin correlations determine the inhomogeneous charge profile, e.g., around a skyrmion or a vortex

Alternative proposals: convert chiral Majorana edge modes or Ising anyons to charged modes via strong coupling to QH edges *and* superconductors

Barkeshli, Berg & Kivelson, Science 2014

Aasen, Mong, Hunt, Mandrus & Alicea, PRX 2020

Example: Hubbard model

Bulaevski et al., PRB 2008
Khomskii, J. Phys. Cond. Matt. 2010

2D Hubbard model at half-filling and strong coupling

$$H = -\sum_{j,k}\left(t_{jk}c_j^+ c_k + h.c.\right) + U\sum_j\left(n_{j\uparrow} - \frac{1}{2}\right)\left(n_{j\downarrow} - \frac{1}{2}\right)$$

→ SU(2) invariant Heisenberg S=1/2 low-energy model

Low-energy form of electron density operator:

$$\hat{n}_j = 1 + \delta\hat{n}_j = 1 + \sum_{kl}\delta\hat{n}_{j,(kl)}$$

sum over site triangles (jkl)

$$\delta\hat{n}_{j,(kl)} = \frac{8t_{jk}t_{kl}t_{lj}}{U^3}\left(\vec{S}_j\cdot\vec{S}_k + \vec{S}_j\cdot\vec{S}_l - 2\vec{S}_k\cdot\vec{S}_l\right)$$

$$\rho_j = -e\left\langle\delta\hat{n}_j\right\rangle$$

local charge imbalance depends on spin-spin correlations!

Spin-orbit assisted Mott insulator

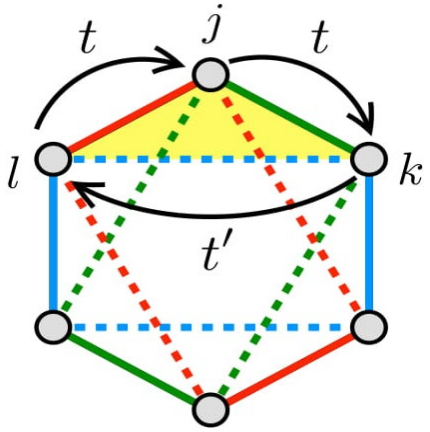
In Kitaev materials, e.g. α-$RuCl_3$: strong spin orbit couplings → SU(2) invariance absent

Start from Hubbard-Kanamori model for d^5 electrons of Ru^{3+} ions in edge-sharing octahedral environment

$$H = H_0 + \sum_j H_{at}^{(j)}$$

Orbital & bond dependent hopping:

$$H_0 = t \sum_{\langle ij \rangle_\gamma} d_{i\alpha}^+ d_{j\beta} + t' \sum_{\langle\langle ij \rangle\rangle_\gamma} d_{i\alpha}^+ d_{j\beta} + h.c.$$



real positive t and t‘ describe transitions between t_{2g} orbitals (xy,yz,zx) ↔ index α=(z,x,y)
after integrating over *p*-orbitals at ligand sites (Cl)

Jackeli & Khaliullin, PRL 2009

Atomic on-site term

effective angular momentum L=1 of t_{2g} orbitals

local electrostatic potential

$$H_{at}^{(j)} = \frac{U-3J_H}{2}\left(N_j-5\right)^2 - 2J_H\vec{S}_j^2 - \frac{J_H}{2}\vec{L}_j^2 - \lambda_{so}\vec{S}_j\cdot\vec{L}_j - eV_0N_j - \vec{h}\cdot\vec{S}_j$$

spin-orbit coupling

magnetic Zeeman field

$RuCl_3$ parameter estimates:

U=2.4 eV, Hund coupling J_H=0.4 eV,

t=0.16 eV, t‘=0.06 eV, $\lambda_{so}\approx$0.15 eV $\longrightarrow$ $t, t' \leq \lambda_{so} < U, J_H$

$\longrightarrow$ effective spin-1/2 description after projecting to low-energy space: single spin-orbital entangled hole per site $j=\frac{1}{2}$

Canonical transformation: eliminate all terms leaving low-energy space (up to desired order)

$$\tilde{H} = e^S H e^{-S}, \tilde{O} = e^S O e^{-S}$$

Local charge imbalance operator

Second order expansion → Kitaev model with ferromagnetic coupling (plus next-nearest Kitaev coupling)

Jackeli & Khaliullin, PRL 2009

$$K = \frac{8 J_H t^2}{3U\left(U - J_H\right)\left(U - 3J_H\right)} \approx 5meV$$

Third-order expansion → local charge operator = sum over site triangles with bond type (αβγ) for pair (jk,jl,kl):

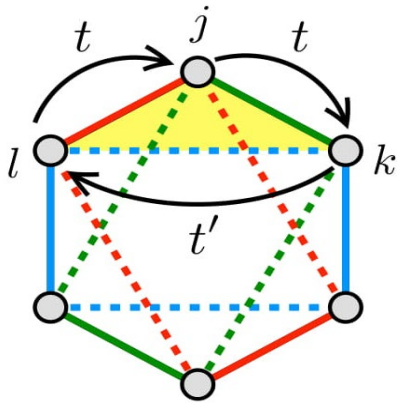
$$\delta\hat{n}_{j,(kl)} = A_1 S_k^\alpha S_j^\alpha + A_2 S_k^\beta S_j^\beta + A_3 S_k^\gamma S_j^\gamma + A_1 S_l^\beta S_j^\beta + A_2 S_l^\alpha S_j^\alpha$$
$$+ A_3 S_l^\gamma S_j^\gamma - 2A_1 S_k^\alpha S_l^\alpha - \left(A_2 + A_3\right)\left[S_k^\alpha S_l^\alpha + S_k^\beta S_l^\beta\right]$$

$$\begin{pmatrix} A_1 \\ A_2 \\ A_3 \end{pmatrix} = \frac{4t' t^2 J_H}{9\left(1-\eta\right)^3\left(1-3\eta\right)^3 U^2} \begin{pmatrix} 3 - 10\eta + 11\eta^2 \\ 5 - 20\eta + 21\eta^2 \\ -5 + 18\eta - 17\eta^2 \end{pmatrix} \qquad \eta = J_H / U$$

For $RuCl_3$ parameters:

$$A_1 \approx 1.86 \cdot 10^{-4}$$

Polarization profile



To leading order in $t'/U << 1$:

local charge imbalance follows

from eigenstates of pure (t'=0) Kitaev model:

$$\rho_j = +e\left\langle \Psi \left| \delta\hat{n}_j \right| \Psi \right\rangle = eA_1 \sum_{kl} \left(\left\langle S_k^\alpha S_j^\alpha \right\rangle + \left\langle S_l^\beta S_j^\beta \right\rangle - 2\left\langle S_k^\gamma S_l^\gamma \right\rangle \right)$$

→ we need spin-spin correlations for Kitaev model

→ can be obtained in exact form!

Ground state (no Z_2 vortices):

Translation invariance → $\rho_j = 0$

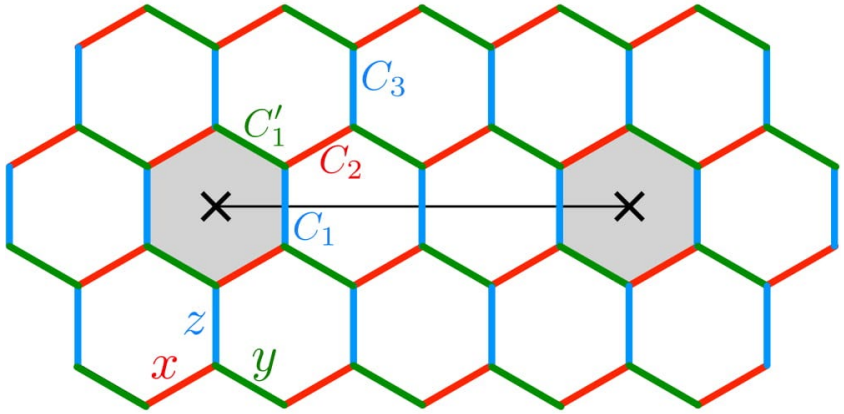
→ minimal case with nontrivial polarization has two vortices

Spin correlations in Kitaev model

Baskaran et al., PRL 2007

Exact spin correlations computable for arbitrary eigenstate of Kitaev model: **Ultra-local form**

$$\left\langle \Psi \left| S_j^\alpha S_l^\beta \right| \Psi \right\rangle = \frac{1}{4} C_{\langle jl \rangle_\alpha} \delta^{\alpha\beta} \qquad C_{\langle jl \rangle_\alpha} = -u_{\langle jl \rangle_\alpha} \left\langle M(G) \left| i c_j c_l \right| M(G) \right\rangle$$



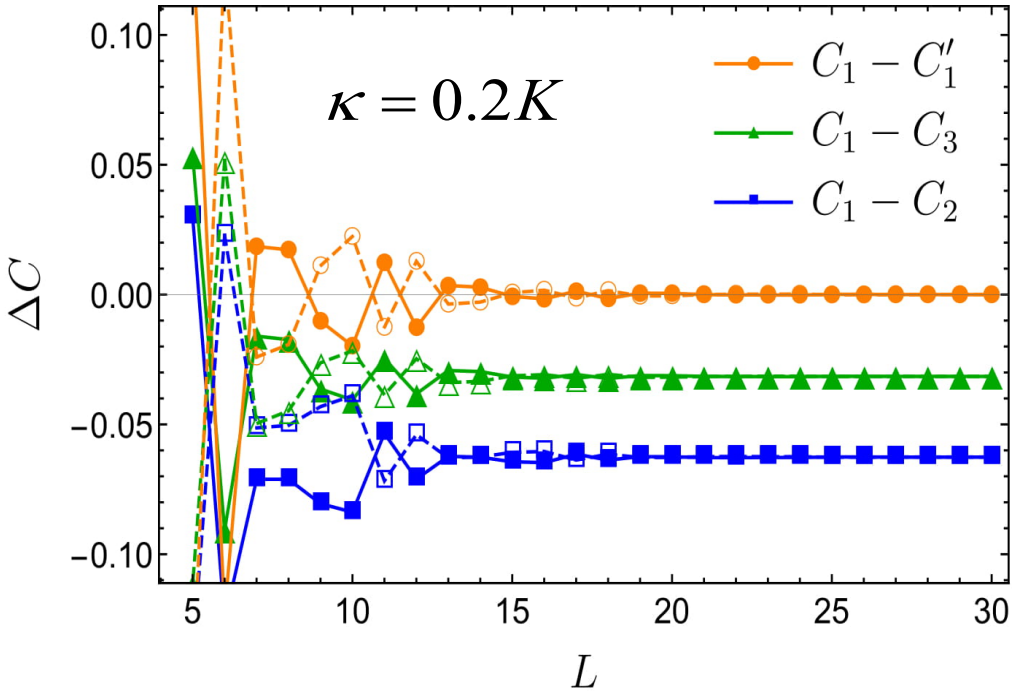
→ numerical diagonalization of LxL Kitaev lattice (p.b.c.) for given vortex configuration

NB: MZM operators for two vortices allow for two parity sectors
But: global fermion parity rule fixes parity sector

Pedrocchi, Chesi & Loss, PRB 2011
Zschocke & Voijta, PRB 2015

Spin correlations near Z_2 vortex

String connecting two vortices is gauge dependent (unphysical)
→ Spin correlations near given vortex are isotropic with all other vortices far away → $C_1' = C_1$



Consider largest possible distance between vortices

$$d = \lfloor (L-1)/2 \rfloor$$

→ thermodynamic limit reached for L>20

Here we find, e.g.,

$$C_1 - C_3 \approx -0.0315$$

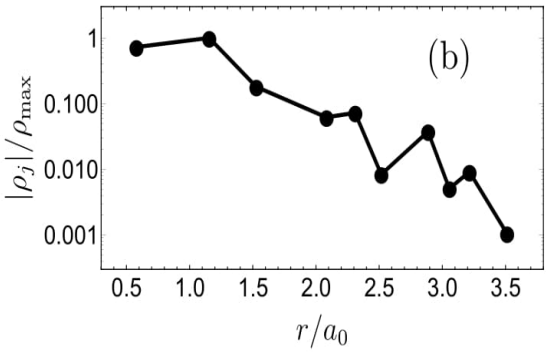
Charge density near Z_2 vortex

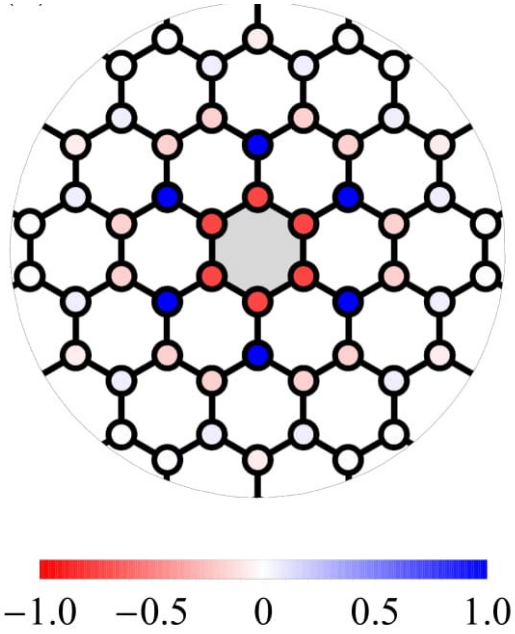
For sites around vortex: Sum over all triangles gives

$\rho_j = eA_1\left(C_1 - C_3\right)$ Here: $\rho_j \approx -5.86 \cdot 10^{-6} e$

Radially isotropic charge profile:

- **Friedel like oscillations**
- Largest charge imbalance on second „ring“, $\rho_{\max} \approx 8.09 \cdot 10^{-6} e$
- Exponential decay at large r
- Overall charge neutrality $\sum_j \rho_j = 0$





→ **small but measurable induced charges (STM)**

Electrostatic potential of vortex

Sum over all lattice sites: $V_{vortex}(\vec{r}) = \sum_j \frac{\rho_j}{|\vec{r} - \vec{R}_j|}$

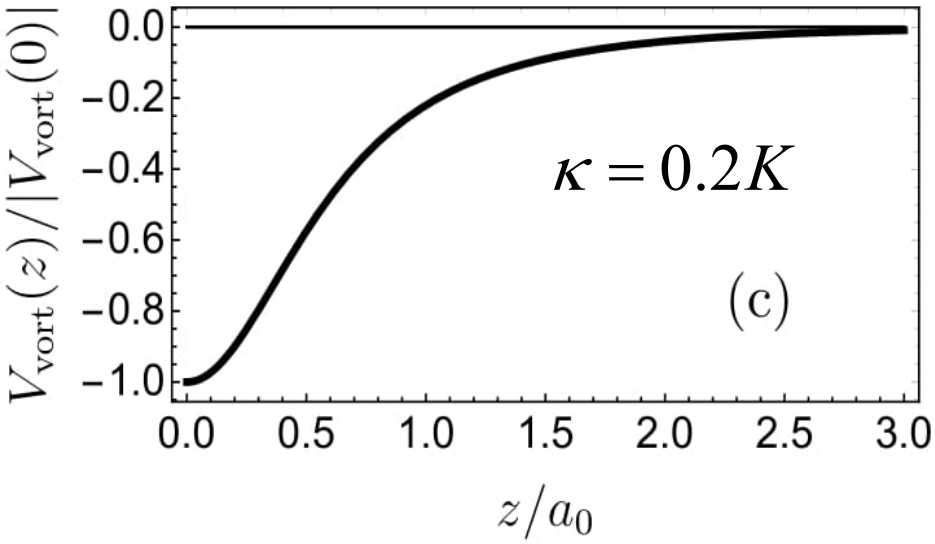
Vortex-induced polarization generates **quadrupolar electrostatic potential,** most pronounced along perpendicular (z) direction.

For large z:

$$V_{vortex}(z) \propto -\frac{1}{|z|^3}$$

For $RuCl_3$ parameters:

$$V_{vortex}(0) \approx -0.118 meV$$



→ **detectable by available AFM techniques**

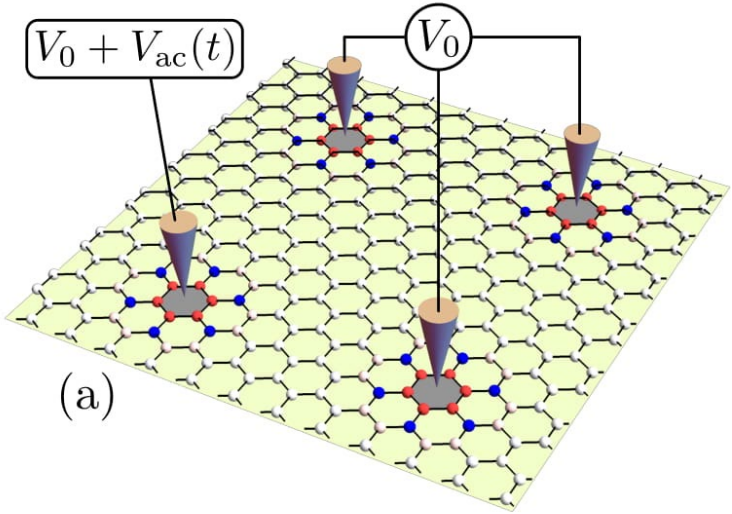
Manipulating vortices: STM/AFM tips

Coupling of local electric fields to Ising anyons?
Consider voltage-biased AFM/STM probe tip above hexagon center → approximate potential by V_0 on adjacent sites and zero otherwise

Include potential in atomic Hamiltonian → projection gives again Kitaev model with renormalized $K(V_0)$ at bonds to second ring

$$K(V_0) = \frac{1+\nu\xi_0^2}{(1-\xi_0^2)(1-\nu^2\xi_0^2)} K \geq K$$

$$\nu = \frac{U-3J_H}{U-J_H} \qquad \xi_0 = \frac{eV_0}{U-3J_H}$$



Projection requires $|\xi_0| < 1$

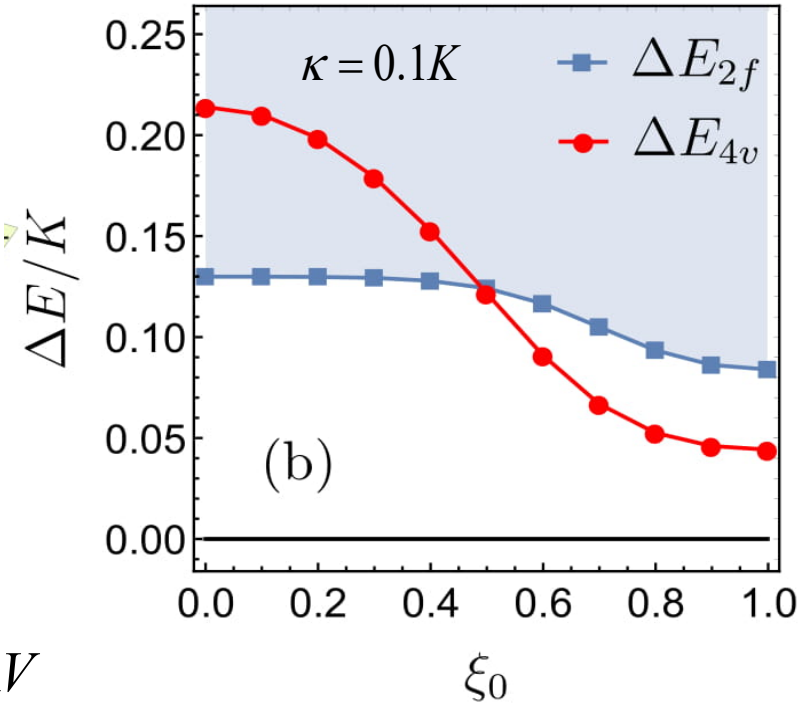
Trapping and moving a vortex

Consider four well separated probe tips at same voltage:
Renormalized Kitaev model still solvable!

Energy gap from zero-vortex g.s.

to four-vortex g.s.: ΔE_{4v}

Gap from g.s. to two-fermion continuum (no vortices): ΔE_{2f}



Vortex under tip gains **stabilization energy**

$$E_v = \Delta E_{4v}(0) - \Delta E_{4v}(V_0) \approx 0.5meV$$

for $\xi_0 = 0.5 \leftrightarrow V_0 \approx 1V$

Vortex attracted to position under tip → trap & adiabatically move vortices

Charge imbalance revisited

If tip-induced voltage breaks site symmetry on a bond:

local charge imbalance appears already at second order (and t‘=0) → larger values!

$$\delta\hat{n}_j = -\frac{8t^2 eV_0}{3U^3} - \frac{16t^2 J_H eV_0}{U^3}\hat{Q}_j,$$

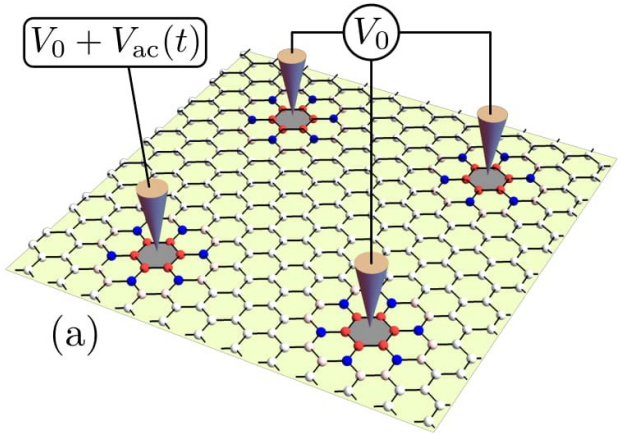
$$\hat{Q}_j \equiv \sum_{\langle jl\rangle_\alpha} S_j^\alpha S_l^\alpha = -\frac{i}{4}\sum_{\langle jl\rangle_\alpha} u_{\langle jl\rangle_\alpha} c_j c_l$$

→ **tip voltage couples to two-spin Q operator**

→ matrix elements of Q can couple ground state to excited states at fixed vortex number (unlike single-spin operators!)

Towards MZM detection

Four-tip setup with 4 vortices:
two-fold g.s. degeneracy due to MZMs
for large distance d (fermion parity rule!)



On tip 1: add weak AC voltage

$$V_{ac}(t) = V_1 \cos(\omega t) \qquad V_1 << V_0$$

Energy absorption probability (Fermi golden rule):

$$P(\omega) = \left(\frac{6e^2 V_0 V_1 K}{(U - 3J_H)^2 U^2} \right)^2 \mathrm{A}(\omega)$$

$|\Psi_n\rangle$: unperturbed (V_1=0) eigenstates to energy E_n

$$\mathrm{A}(\omega) = \frac{2\pi}{\hbar} \sum_n \left| \langle \Psi_n | \hat{Q}_1 | \Psi_0 \rangle \right|^2 \delta(\omega - E_n + E_0)$$

can be measured by spectroscopic techniques!

Rugar et al., Nature 2004
Baumann et al., Science 2015

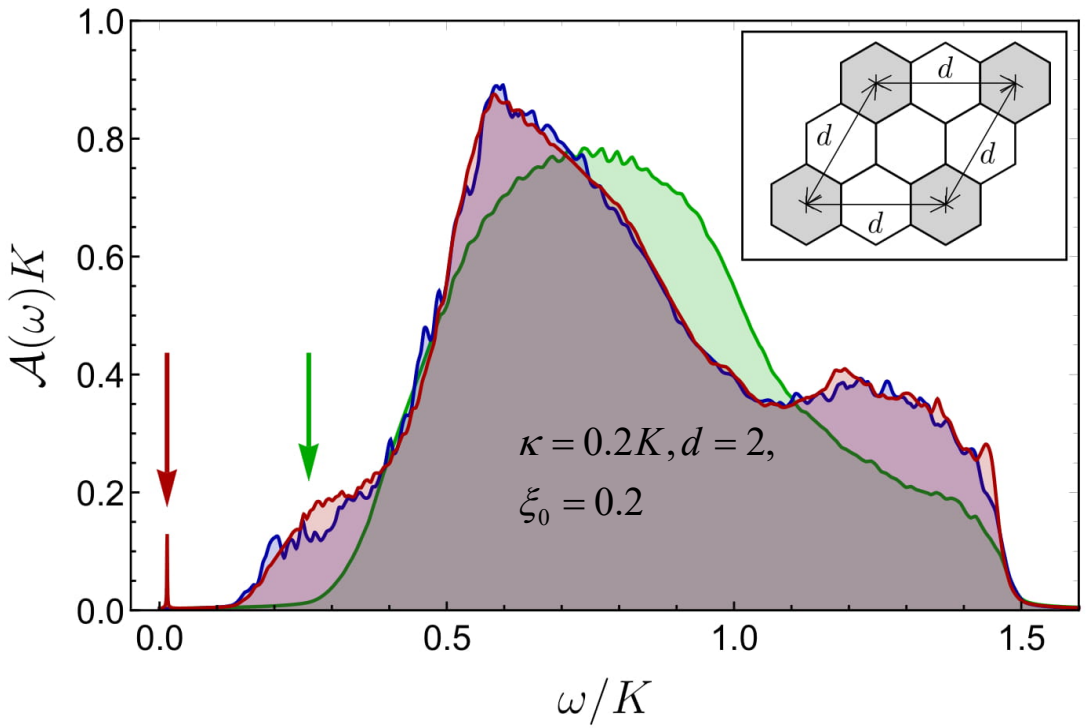
Energy absorption spectrum

MZM peak: $\omega = \varepsilon_M(d)$

- **only for N_v=4**
- hybridization energy $\varepsilon_M(d)$ & peak weight vanish for d→∞

Number of vortices below tips:

N_v=0
N_v=2 (incl. tip 1)
N_v=4



Continuum threshold for N_v=2,4: one bulk fermion + zero mode →

$$\omega \approx \Delta E_f$$

for N_v=0: 2 bulk fermions

→ $\omega = \Delta E_{2f} \approx 2\Delta E_f$

→ another MZM signature via threshold energy

Conclusions

- **Introduction**
  - Kitaev honeycomb model in magnetic field: Non-Abelian chiral spin liquid
  - Ising anyon: Z_2 vortex with Majorana zero mode (MZM)
  - Kitaev materials, e.g., α-$RuCl_3$
  - Electric polarization effects in Mott insulators

- **Local charge imbalance in Kitaev materials near Z_2 vortices**
  Charge density profile around vortex is related to spin correlations
  → generates quadrupolar electric field

- **Trapping & moving vortices by STM/AFM tips**

- **Energy absorption spectroscopy**
  ⟶ detection of MZMs in Kitaev spin liquids

R. G. Pereira and R. Egger, PRL (in press), arXiv:2008.05334

THANK YOU FOR YOUR ATTENTION!