Perturbing the U(1) Dirac Spin Liquid State in Spin-1/2 kagome Raman scattering, magnetic field, and hole doping



Acknowledgments



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Outline

- 1 The Spin-1/2 Kagome Lattice
- **2** Derivation and Properties of the U(1) DSL State
- 3 Raman Scattering
- 4 External Magnetic Field
- **5** Hole Doping

Reference: Ran, Ko, Lee, & Wen, PRL **102**, 047205 (2009) Ko, Lee, & Wen, PRB **79**, 214502 (2009) Ko, Liu, Ng, & Lee, PRB **81**, 024414 (2010)

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The nearest-neighbor antiferromagnetic Heisenberg model is highly frustrated on the kagome lattice.

- kagome lattice = 2D lattice of corner-sharing triangles.
- Simple Néel order does not work well on the kagome lattice.
- Classical ground states: $\sum_{i \in \triangle} \mathbf{S}_i = 0$.

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In the quantum case, singlet formation is possible and may be favored.

• 1D chain:

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Two major classes of singlet states: valence bond solids (VBS) and spin liquid (SL).

- In a VBS, certain singlet bonds are preferred, which results in a symmetry-broken state.
- In a SL, different bond configurations superpose, which results in a state that breaks no lattice symmetry.
- A VBS state generally has a spin gap, while a SL state can be gapped or gapless.



VS.



For S=1/2 kagome, the leading proposals are the 36-site VBS and the U(1) Dirac spin liquid.

- The 36-site VBS pattern is found in series expansion [Singh and Huse, PRB **76**, 180407 (2007)] and entanglement renormalization [Evenbly and Vidal, arXiv:0904.3383].
- From VMC, the U(1) Dirac spin liquid (DSL) state has the lowest energy among various SL states, and is stable against small VBS perturbations [e.g., Ran et al., PRL 98, 117205 (2007)].
- Exact diagonalization: initially found small (~ J/20) spin gap; now leaning towards a gapless proposal [Waldtmann *et al.*, EPJB 2, 501 (1998); arXiv:0907.4164].





Experimental realization of S = 1/2 kagome: Herbertsmithite ZnCu₃(OH)₆Cl₂.

- Herbertsmithite: layered structure with Cu forming an AF kagome lattice.
- Caveats: Zn impurities and Dzyaloshinskii– Moriya interactions.
- Experimental Results [e.g., Helton et al., PRL 98, 107204 (2007); Bert et al., JP:CS 145, 012004 (2009)]:
 - Neutron scattering: no magnetic order down to 1.8 K.
 - μ SR: no spin freezing down to 50 μ K.
 - Heat capacity: vanishes as a power law as $T \rightarrow 0$.
 - Spin susceptibility: diverges as $T \rightarrow 0$.
 - NMR shift: power law as $T \rightarrow 0$.



Research motivation: Deriving further experimental consequences of the U(1) DSL state.

- All experiments point to a state without magnetic order. But more data is needed to tell if it is a VBS state or a SL state, and which VBS/SL state it is.
- Without concrete theory, the experimental data are hard to interpret.
- Without concrete theory, unbiased theoretical calculations are difficult.
- The *U*(1) Dirac spin-liquid state is a theoretically interesting exotic state of matter.

Thus, our approach: Assume the DSL state and consider further experimental consequences.

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$$H_{tJ} = \sum_{\langle ij \rangle} J\left(\mathbf{S}_i \cdot \mathbf{S}_j - \frac{1}{4}n_i n_j\right) - t\left(c_{i\sigma}^{\dagger}c_{j\sigma} + h.c.\right); \qquad \sum_{\sigma} c_i^{\dagger}c_i \leq 1$$

• Apply the slave boson decomposition [Lee *et al.*, RMP 78, 17 (2006)]: $\mathbf{S}_i = \frac{1}{2} \sum_{\alpha,\beta} f_{i\alpha}^{\dagger} \tau_{\alpha,\beta} f_{i\beta} ; \quad c_{i\sigma}^{\dagger} = f_{i\sigma}^{\dagger} h_i ; \quad f_{i\uparrow}^{\dagger} f_{i\uparrow} + f_{i\downarrow}^{\dagger} f_{i\downarrow} + h_i^{\dagger} h_i = 1$

- Decouple four-operator terms by a Hubbard–Stratonovich transformation, with the following ansatz: $\chi_{ij} \equiv \sum_{\sigma} \langle f_{i\sigma}^{\dagger} f_{j\sigma} \rangle = \chi e^{i\alpha_{ij}}; \quad \Delta_{ij} \equiv \langle f_{i\uparrow}^{\dagger} f_{j\downarrow} - f_{i\downarrow}^{\dagger} f_{j\uparrow} \rangle =$
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Deriving the U(1) DSL state: Emergent gauge field

$$\begin{aligned} H_{\rm MF} &= \sum_{i\sigma} f^{\dagger}_{i\sigma} (i\alpha^{i}_{0} - \mu_{F}) f_{i\sigma} - \frac{3\chi J}{8} \sum_{\langle ij \rangle, \sigma} (e^{i\alpha_{ij}} f^{\dagger}_{i\sigma} f_{j\sigma} + h.c.) \\ &+ \sum_{i} h^{\dagger}_{i} (i\alpha^{i}_{0} - \mu_{B}) h_{i} - t\chi \sum_{\langle ij \rangle} (e^{i\alpha_{ij}} h^{\dagger}_{i} h_{j} + h.c.) \end{aligned}$$

- The α field is an emergent gauge field, corresponding to gauge symmetry $f^{\dagger} \mapsto e^{i\theta} f^{\dagger}, h \mapsto e^{-i\theta} h$.
- At the lattice level *α* is a *compact* gauge field (i.e., monopoles are allowed).
- But with Dirac fermions, the system can be in a deconfined phase (i.e., monopoles can be neglected) [Hermele *et al.*, PRB **70**, 214437 (2004)].

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Deriving the U(1) DSL state: Band Structure

$$\begin{split} H_{\rm MF} &= \sum_{i\sigma} f^{\dagger}_{i\sigma} (i\alpha^{i}_{0} - \mu_{F}) f_{i\sigma} - \frac{3\chi J}{8} \sum_{\langle ij \rangle, \sigma} (e^{i\alpha_{ij}} f^{\dagger}_{i\sigma} f_{j\sigma} + h.c.) \\ &+ \sum_{i} h^{\dagger}_{i} (i\alpha^{i}_{0} - \mu_{B}) h_{i} - t\chi \sum_{\langle ij \rangle} (e^{i\alpha_{ij}} h^{\dagger}_{i} h_{j} + h.c.) \end{split}$$

- Neglecting fluctuation of *α*, spinons and holons are decoupled.
- Mean-field ansatz for SL state can be specified by pattern of α flux.
- U(1) Dirac spin liquid state: π flux per \bigcirc and 0 flux per \triangle .
- π flux \implies unit cell doubled in band structures.



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/
$$t_{\rm eff} = +t$$
 ; / $t_{\rm eff} = -t$



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Perturbing the U(1) DSL State

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Properties of the U(1) DSL state: Thermodynamics and correlation

- At low energy, the U(1) DSL state is described by QED₃.
 - i.e., gauge field coupled to Dirac fermions in (2+1)-D.
- Thermodynamics of the U(1) DSL state is dominated by the spinon Fermi surface.
 - Zero-field spin susceptibility: $\chi(T) \sim T$.
 - Heat capacity: $C_V(T) \sim T^2$.
- U(1) DSL state is "quantum critical" many correlations decay algebraically [Hermele *et al.*, PRB **77**, 224413 (2008)].
 - Emergent SU(4) symmetry among Dirac nodes ⇒ different correlations can have the same scaling dimension.



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Outline

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3 Raman Scattering

4 External Magnetic Field

5 Hole Doping

Raman scattering in Mott-Hubbard system: the Shastry–Shraiman formulation

- Raman scattering = inelastic scattering of photon.
 - Good for studying excitations of the system.
 - Probe only excitations with $\mathbf{q} \approx 0$.
- We are concerned with a half-filled Hubbard system, in the regime where $|\omega_i \omega_f| \ll U$ and $\omega_i \approx U$.
 - Both initial and final states are spin states.
 - \implies T-matrix can be written in terms of spin operators.
 - Intermediate states are dominated by the sector where $\sum_{i} n_{i\uparrow} n_{i\downarrow} = 1$.
 - The T-matrix can be organized as an expansion in $t/(U \omega_i)$ [Shastry & Shraiman, IJMPB 5, 365 (1991)].



$$T^{(2)} \sim rac{t^2}{U-\omega_i} \mathbf{S}_i \cdot \mathbf{S}_j + \dots$$

Spin-chirality terms in the Shastry-Shraiman formulation

• Because of holon-doublon symmetry, there is no t^3 order contribution.



• For the square and triangular lattice, there is no t⁴ order contribution to spin-chirality because of a non-trivial cancellation between 3-site and 4-site pathways.



• But such cancellation is absent in the kagome lattice.

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Raman T-matrix for the kagome geometry

• For kagome geometry, the Raman T-matrix decomposes into 3 irreps:

 $T = T(A_{1g})(\bar{x}x + \bar{y}y) + T(E_g^{(1)})(\bar{x}x - \bar{y}y) + T(E_g^{(2)})(\bar{x}y + \bar{y}x) + T(A_{2g})(\bar{x}y - \bar{y}x)$

• To lowest order in inelastic terms,

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- $\mathbf{S}_i \cdot \mathbf{S}_j \sim f^{\dagger} f f^{\dagger} f$ and $\mathbf{S}_i \cdot (\mathbf{S}_j \times \mathbf{S}_k) \sim f^{\dagger} f f^{\dagger} f f^{\dagger} f$
 - \implies contributions from spinon-antispinon pairs.
 - \implies continuum of signal $I_{\alpha}(\Delta \omega) = |\langle f | O_{\alpha} | i \rangle|^2 \text{DOS}(\Delta \omega).$
 - At low energy, one-pair states dominates.
 - For Dirac node, $\text{DOS}_{1pair} \sim \mathcal{E}$, and matrix element is suppressed in E_g and A_{1g} , but not in A_{2g} .

 $\implies \mbox{Spinon-antispinon pairs contribute } I_{A_{2g}}(\Delta\omega) \sim \mathcal{E} \mbox{ and } I_{E_g/A_{1g}}(\Delta\omega) \sim \mathcal{E}^3 \mbox{ at low energy.}$

- However, an additional collective excitation is available in A_{2g}: S_i · S_j × S_k ~ iχ³ exp(i ∮_Δ α · dℓ) + h.c. ~ χ³ ∫∫_Δ b d²x ⇒ I_{A_{2g}} ~ ⟨Φ_bΦ_b⟩ + ... ~ q²⟨αα⟩ + ... (Recall that ⟨f_i[†]f_j⟩ ~ χ exp(iα_{ij}))
 - In our case (QED₃ with Dirac fermions), turns out that $\langle \alpha \alpha \rangle \sim 1/\omega$ when $\mathbf{q} \approx 0$ [loffe & Larkin, PRB **39**, 8988 (1989)].
- Analogy: plasmon mode vs. particle-hole continuum in normal metal.

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Experimental Comparisons: Some qualitative agreements

- Wulferding and Lemmens [unpublished] have obtained Raman signal on herbertsmithite.
 - At low T, data shows a broad background with a near-linear piece at low-energy.
 - Roughly agree with the theoretical picture presented previously.



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External magnetic field and the formation of Landau levels

- In Mott systems, B-field causes only Zeeman splitting.
 - This induces spinon and antispinon pockets near the Dirac node.
- However, with the emergent gauge field α, Landau levels can form spontaneously.
- From VMC calculations,

 $\Delta e_{FP}^{Prj} pprox 0.33(2)B^{3/2} + 0.00(4)B^2$ $\Delta e_{LL}^{Prj} pprox 0.223(6)B^{3/2} + 0.03(1)B^2$

$$\begin{split} \Delta e^{LL}_{MF} &< \Delta e^{FP}_{MF} \text{ to leading order in } \Delta n \\ \Longrightarrow \text{ Landau level state is favored.} \end{split}$$



- *b* is an emergent gauge field
 ⇒ its strength can fluctuate in space.
- The fluctuation of b is tied to the fluctuation of S_z density.
- In long-wavelength limit, energy cost of b fluctuation $\rightarrow 0$
 - \implies The system has a gapless mode!
 - Derivative expansion \implies linear dispersion.
- Other density fluctuations and quasiparticle excitations are gapped ⇒ gapless mode is unique.
- Mathematical description given by Chern–Simons theory.



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Gapless mode and XY-ordering

- Recap: we found a single linearly-dispersing gapless mode, which looks like...
- A Goldstone boson! And indeed it is.
 - Corresponding to this Goldstone mode is a spontaneously broken XY order.
- Analogy:
 - Superfluid: $\hat{\psi} = \sqrt{\hat{\rho}} e^{-i\hat{\theta}}$, $[\hat{\rho}, \hat{\theta}] = i$, gapless ρ fluctuation \implies ordered (SF) phase;
 - XY model: $\hat{S^+} = e^{i\hat{\theta}}$, $[\hat{S_z}, \hat{\theta}] = i$, gapless S_z fluctuation \implies XY ordered phase.
- VMC found the $\mathbf{q} = \mathbf{0}$ order.
- S^+ in XY model $\sim V^{\dagger}$ in QED₃ \implies in-plane magnetization $M \sim B^{\gamma}$. (V^{\dagger} monopole operator, γ its scaling dimension)



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Recap: Band structure



Hole doping and formation of Landau levels

- Doping can in principle be achieved by substituting CI with S.
- In slave-boson picture, hole doping introduces holons and antispinons.
- As before, an emergent *b* field can open up Landau levels in the spinon and holon bands.
 - At mean-field, $\Delta E_{\rm spinon} \sim -B^{3/2}$ while $\Delta E_{\rm holon} \sim B^2$ \implies LL state favored.
 - At mean-field, b optimal when antispinons form $\nu=-1~{\rm LL}$ state
 - \implies holons form $\nu=1/2$ Laughlin state.



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Hole Doping

Charge fluctuation, Goldstone mode, and superconductivity

- *b* fluctuation \sim holon density fluctuation \sim charge density fluctuation.
- Long-wavelength *b* fluctuation cost $\mathcal{E} \searrow 0$ if real EM-field is "turned off." \implies a single linearly-dispersing mode
 - \sim Goldstone boson.
- This time the Goldstone boson is eaten up by the EM-field to produce a superconductor.
 - This superconducting state breaks *T*-invariance.
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- Quasiparticles are bound states of semions in holon sectors and/or fermions in spinon sector.
 - For finite energy, bound states must be neutral w.r.t. the gapless mode.
- There are two types of "elementary" quasiparticles:
 - 1 semion-antisemion pair in holon sector;
 - 2 spinon-holon pair
 - (\sim Bogoliubov q.p. in conventional SC).
- All other quasiparticles can be built from these elementary ones.
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Crystal momenta of quasiparticle—projective symmetry group study

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 $\mathbf{k} = \mathbf{k}$ of spinon-holon pairs

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Conclusions

- The *U*(1) Dirac spin-liquid state possess many unusual properties and may be experimentally realized in herbertsmithite.
- The Raman signal of the DSL state has a broad background (contributed by spinon-antispinon continuum) and a $1/\omega$ singularity (contributed by collective [gauge] excitations).
- Under external magnetic field, the DSL state forms Landau levels, which corresponds to a XY symmetry broken state with Goldstone boson corresponding to S_z density fluctuation.
- When the DSL state is doped, an analogous mechanism give rise to an Anderson-Higgs scenario and hence superconductivity.
 - But minimal vortices carry *hc*/4*e* flux and the system contains exotic quasiparticle having semionic mutual statistics.

Reference: Ran, Ko, Lee, & Wen, PRL **102**, 047205 (2009) Ko, Lee, & Wen, PRB **79**, 214502 (2009) Ko, Liu, Ng, & Lee, PRB **81**, 024414 (2010)

Appendix

(a.k.a. hip pocket slides)

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Perturbing the U(1) DSL State

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Comparison of ground-state energy estimate

Method	Max. Size	Energy	State	Ref.
Exact Diag.	36	-0.43		[1]
DMRG	192	-0.4366(7)	SL	[2]
VMC	432	-0.42863(2)	U(1) Dirac SL	[3]
Series Expan.	—	-0.433(1)	36-site VBS	[4]
Entang. Renorm.	—	-0.4316	36-site VBS	[5]

- Waldtmann *et al.*, EPJB **2**, 501 (1998)
 Jiang *et al.*, PRL **101**, 117203 (2008)
- [3] Ran *et al.*, PRL **98**, 117205 (2007)
- [4] Singh and Huse, PRB 76, 180407 (2007)
- [5] Evenbly and Vidal, arXiv:0904.3383

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Appendix

Magnified band structure of DSL state, with scales





Brillouin zone

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Perturbing the U(1) DSL State

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Appendix

Raman signals contributed by spinon-antispinon: full scale



Perturbing the U(1) DSL State

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Chern-Simons description: B-field case

• For the B-field case, introduce two species of gauge fields to describe the current of up/down spins:

$$J^{\mu}_{\pm}=rac{1}{2\pi}\epsilon^{\mu
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• The Lagrangian in terms of α and a_{\pm} :

$$\mathcal{L} = \pm \frac{1}{4\pi} \epsilon^{\mu\nu\lambda} \mathbf{a}_{\pm,\mu} \partial_{\nu} \mathbf{a}_{\pm,\lambda} + \frac{1}{2\pi} \epsilon^{\mu\nu\lambda} \alpha_{\mu} \partial_{\nu} \mathbf{a}_{\pm,\lambda} + \dots$$

- higher derivative terms (e.g., Maxwell term $\sim \partial a \partial a$ for a_{\pm}) omitted
- \mathcal{L} yields the correct equation of motion $J^{\mu}_{\pm} = \mp \frac{1}{2\pi} \epsilon^{\mu\nu\lambda} \partial_{\nu} \alpha_{\lambda}$
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- Dynamics of c_0 is driven by Maxwell term \implies linearly dispersing.

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In the doped case:

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where
$$\mathbf{c} = [\alpha; a_1, \dots a_4, a_5, a_6; b_1, \dots b_4]$$

spinon spinon* holon

- *K* describes the self-dynamics of the system; has null vector $\mathbf{p}_0 = [2; -2, \dots, -2, 2, 2; 1, \dots, 1]$ corresponding to gapless mode c_0
- $\mathbf{q} = [0; 0 \dots 0, 0, 0; 1, \dots, 1]$ is the "charge vector."
- ℓ is an integer vector with 0 α -component and characterizes vortices.
- Varying \mathcal{L} w.r.t. c_0 gives $B = -\frac{2\pi}{e} \frac{\ell \cdot \mathbf{p}_0}{\mathbf{q} \cdot \mathbf{p}_0} j_V^0 = -\frac{2n\pi}{4e} j_V^0$

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- ℓ is an integer vector with 0 α -component and characterizes vortices.
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In the doped case:

$$\mathcal{L} = \underbrace{-\frac{1}{4\pi} \epsilon^{\mu\nu\lambda} \mathbf{c}_{\mu}^{T} K \partial_{\nu} \mathbf{c}_{\lambda}}_{\text{self dynamics}} + \underbrace{\frac{e}{2\pi} \epsilon^{\mu\nu\lambda} (\mathbf{q} \cdot \mathbf{c}_{\mu}) \partial_{\nu} A_{\lambda}}_{\text{self dynamics}} + \underbrace{(\ell \cdot \mathbf{c}_{\mu}) j_{V}^{\mu}}_{\text{vortices}} + \dots$$
where $\mathbf{c} = [\alpha; a_{1}, \dots a_{4}, a_{5}, a_{6}; b_{1}, \dots b_{4}]$
spinon spinon* holon

- *K* describes the self-dynamics of the system; has null vector $\mathbf{p}_0 = [2; -2, \dots, -2, 2, 2; 1, \dots, 1]$ corresponding to gapless mode c_0 .
- $\mathbf{q} = [0; 0 \dots 0, 0, 0; 1, \dots, 1]$ is the "charge vector."
- ℓ is an integer vector with 0 α -component and characterizes vortices.

• Varying
$$\mathcal{L}$$
 w.r.t. c_0 gives $B = -\frac{2\pi}{e} \frac{\ell \cdot \mathbf{p}_0}{\mathbf{q} \cdot \mathbf{p}_0} j_V^0 = -\frac{2n\pi}{4e} j_V^0$

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Quasiparticles and their statistics

$$\mathcal{L} = -rac{1}{4\pi} \epsilon^{\mu
u\lambda} \mathbf{c}_{\mu}^{T} \mathcal{K} \partial_{
u} \mathbf{c}_{\lambda} + rac{e}{2\pi} \epsilon^{\mu
u\lambda} (\mathbf{q} \cdot \mathbf{c}_{\mu}) \partial_{
u} \mathcal{A}_{\lambda} + (\boldsymbol{\ell} \cdot \mathbf{c}_{\mu}) j_{V}^{\mu} + \dots$$

- Vortices with ℓ · p₀ = 0 does not couple to c₀
 ⇒ They can exist when B = 0 and corresponds to quasiparticles.
- When particle ℓ_1 winds around another particle ℓ_2 , the statistical phase $\theta = 2\pi \ell_1^T K_{\perp}^{-1} \ell_2$
 - K_{\perp} is part of K that's $\perp \mathbf{p}_0$.
 - Derived by integrating out all gauge fields having non-zero Chern–Simons term.

• Taking
$$\begin{cases} \ell_1 &= [0; 0, \dots, 0, 0, 0; 1, 0, 0, -1] \\ \ell_2 &= [0; 0, \dots, 0, 0, 0; 1, 0, -1, 0] \\ \theta_{11} &= \theta_{22} = 2\pi \end{cases}$$
, found $\theta_{12} = \pi$

→ Fermions with semionic statistics!

Appendix

The full form of K-matrix for doped case

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Spinon and holon PSG



$$T_{x}[\varphi_{1}^{2}] = \varphi_{4}^{2}$$

$$T_{x}[\varphi_{2}^{2}] = \varphi_{3}^{2}$$

$$T_{x}[\varphi_{3}^{2}] = e^{-\frac{i\pi}{3}}\varphi_{2}^{2}$$

$$T_{x}[\varphi_{4}^{2}] = e^{\frac{i\pi}{3}}\varphi_{1}^{2}$$



$$\begin{split} T_{x}[\eta_{1}] &= e^{\frac{i\pi}{12}}\eta_{2} \\ T_{x}[\eta_{2}] &= e^{\frac{11i\pi}{12}}\eta_{1} \\ T_{x}[\eta_{3}] &= e^{\frac{i\pi}{12}}\eta_{4} \\ T_{x}[\eta_{4}] &= e^{\frac{11i\pi}{12}}\eta_{3} \end{split}$$