Dynamics of 2- and 3-dimensional Quantum Spin Liquids

OAK, September 2015 Kolymbari

Johannes Knolle
TCM Cavendish Lab – University of Cambridge

Collaborators: Roderich Moessner (MPI PKS Dresden)
Dima Kovrizhin (Cambridge U)
John Chalker (Oxford U)
Gia-Wei Chern (Los Alamos NL)
Natalia Perkins (U of Minneapolis)
Brent Perreault (U of Minneapolis)
Fiona Burnell (U of Minneapolis)
Outline

1. Introduction and Motivation
   → New states of matter and fractionalization
   → Spin liquids and dynamics

2. Kitaev’s Honeycomb Model
   → Exact solution
   → 2- and 3-dimensional candidate materials

3. Exact Dynamical Correlation Functions
   → Spin structure factor and the X-ray edge
   → Raman scattering

4. Conclusion
1. Introduction

- Understanding phases of matter and their transitions. Democritus 500 BC
- Phases are described by symmetries and order parameters. Landau 1937

This work
Quantum Spin Liquids

- Frustrated magnets can evade ordering down to $T=0$.
  Anderson 1973

- **QSL**: State of interacting spins that breaks no rotational or translational symmetry and has only short range spin correlations.

- **RVB**: Topologically ordered with fractional excitations.
  Wen 1991

- **Topologically ordered with fractional excitations.**  Wen 1991

---

1. Introduction and Motivation

- FQHE

- Spinons in QSLs
QSL and Dynamics

- Shortage of experimental signatures due to lack of local order.

- Look for signatures of QSL in the **dynamical response**!
  - Inelastic Neutron Scattering
  - Established in 1D
    - Bethe 1931
    - Caux, Tennant 2012

- Poor understanding of strongly interacting systems beyond 1D:
  - No exact results, only numerics or uncontrolled approximations

- Here: **First exact calculation of a dynamical correlation function for a strongly interacting 2D and 3D QSL.**
Main Questions:

How to diagnose a quantum spin liquid in real experiment?
How to probe fractionalized quasiparticles?

Take Home Message:

Signatures of fractionalization are visible in dynamical correlation functions accessible by standard experiments.
2. Kitaev’s Model

- Spin $\frac{1}{2}$ on the honeycomb lattice with strong spin orbit coupling
  Kitaev 2006

$$H = -J_x \sum_{x \text{ links}} \sigma_j^x \sigma_k^x - J_y \sum_{y \text{ links}} \sigma_j^y \sigma_k^y - J_z \sum_{z \text{ links}} \sigma_j^z \sigma_k^z$$

- Large number of conserved quantities, local plaquette operators:
  $$W_p = \sigma_1^x \sigma_2^y \sigma_3^z \sigma_4^x \sigma_5^y \sigma_6^z$$
  $$[H, W_p] = 0$$

- Exactly solvable interacting 2D model!
Exact Solution

- Mapping spins to Majoranas:

\[ \sigma_i^a = i c_i c_i^a, \quad a = x, y, z \]

- Quadratic Hamiltonian in each flux sector:

\[ H = - \sum_{a=x,y,z} J_a \sum_{\langle ij \rangle_a} i c_i \hat{u}_{\langle ij \rangle_a} c_j \text{ with } \hat{u}_{\langle ij \rangle_a} \equiv i c_i^a c_i^a \]

- Spectrum:

- Phase diagram:
Candidate Materials

- Strong spin orbit interaction of late transition metal ions (e.g. Ir).
  - Jackeli, Khaliullin 2009, Singh, Gegenwart 2010

- Recent 3D harmonic honeycomb series for Li$_2$IrO$_3$ and α-RuCl$_3$
3. Dynamical Correlations

- Spin correlation function:

\[
S_{R,R'}^{ab}(t) = \langle MG | \langle G | \sigma_R^a(t) \sigma_{R'}^b(0) | G \rangle | MG \rangle \\
\sigma_i^a \to ic_i \hat{\Pi}^{\left\langle ij \right\rangle,a} \hat{\Pi}^{\left\langle ij \right\rangle,a} 
\]

- Only N.N. spin correlations from flux selection rule

- Dynamics from the perturbed **Majorana sector** with **two extra fluxes** neighboring the N.N. bond

\[
S_{R,R'}^{ab}(t) = -i \langle MG | e^{iH_0 t} c_R e^{-it(H_0+V_a)} c_{R'} | MG \rangle \delta_{a,b} \delta_{\langle R,R' \rangle_{N.N.}} \\
V_a = -J_a 2ic_R c_{R'} 
\]

→ Quadratic **non-equilibrium** problem

Baskaran, PRL 2007
Spin Structure Factor

- Spin correlation function as a **local quantum quench**

\[ S_{ij}^{ab}(t) = -i \langle M_0 | e^{i \hat{H}_0 t} \hat{c}_i e^{-i (\hat{H}_0 + \hat{V}_a) t} \hat{c}_j | M_0 \rangle \delta_{ab} \delta_{ij}, a \]

- Lehmann representation: lowest response and few particle contribution.

\[ S_{ij}^{ab}(\omega) = -i \sum_{\lambda} \langle M_0 | \hat{c}_i | \lambda \rangle \langle \lambda | \hat{c}_j | M_0 \rangle \times \delta [\omega - (E_{\lambda} - E_0)] \delta_{ij}, a \delta_{ab} \]

- Bogoliubov rotation

\[ \hat{b}_\lambda = \sum_q X_{\lambda q}^* \hat{a}_q + Y_{\lambda q}^* \hat{a}_q^\dagger \]

\[ \text{diagonalizes } \hat{H}_0 + \hat{V}_a \]

- \[ |\lambda_0 \rangle = [X^\dagger X]^{1/4} e^{-\frac{1}{2} \hat{a}_q^\dagger X^{*-1} Y^* \hat{a}_q^\dagger} |M_0 \rangle \]

\[ \text{gives the ground state} \]

- Single particle contribution from the s.p. states

\[ |\lambda \rangle = \hat{b}_{\lambda}^\dagger |\lambda_0 \rangle \]

\[ S^{zz}(q = 0, \omega) = \frac{2}{N} \left| \text{det } X \right| \sum_{\lambda, qq'} \delta [\omega - (E_{\lambda}^1 - E_0)] \cos \theta_q X_{q\lambda}^{-1} [X^{-1}]_{\lambda q'}^\dagger \cos \theta_{q'} \]
Results 2D QSL

- Response on the full phase diagram for Abelian and non-Abelian QSL → Salient signatures of fractionalization are visible!
Results Gapless QSL $J_z=J_x=J_y$

- Exact structure factor $S(q=0,\omega)$ for a 2D quantum spin liquid:
  - Flux gap of $\Delta \approx 0.26$ and features of the matter fermion DOS
  - Salient signatures of fractionalization are visible!
Results Gapless Phase $J_z = J_x = J_y$

- Momentum-dependence of $\text{Im} \chi(q, \omega)$ as measured by INS, FM vs AFM

- Broad features in reciprocal space $\rightarrow$ N.N. correlations only
- Results in full phase diagram $\rightarrow$ $\delta$-function in dimer limit!
Results Gapless Phase $J_z = J_x = J_y$

- Powder Average as measured in INS for AFM Kitaev coupling

\[ \frac{d\sigma}{dE} \propto \frac{q}{q_{\text{in}}} \int d\theta |f(q)|^2 \left[ (1 - \cos^2 \theta)S^{xx}(q, \omega) + (1 - \sin^2 \theta)S^{yy}(q, \omega) + S^{zz}(q, \omega) \right] \]
Exact Solution and the X-Ray Edge

- In terms of complex matter fermions:

\[ f_r = \frac{1}{2}(\hat{c}_{Ar} + i\hat{c}_{Br}) \]

\[ S_{A0B0}^{zz}(t) = i[g(t, 0) + g(0, t)] \]

\[ g(t, 0) = -i\langle M_0|T\left\{f_0(t)f_0^*(0)\exp\left[-i\int_0^t \hat{V}_z(\tau)d\tau\right]\right\}|M_0\rangle \]

with \( \hat{V}_z(t) = v\left[f_0^*(t)f_0(t) - \frac{1}{2}\right] \)

- Local quench equivalent to a X-ray edge problem!

Nozieres, DeDominicis 1969
Exact Solution

- Problem reduces to solving a **singular integral equation**.

\[ G_c(t, t') = G_0(t, t') - 4J_z \int_{t'}^t dt_1 G_0(t, t_1) G_c(t_1, t'). \]

- Solved for \( t \to \infty \) for a Fermi liquid with \( G_0(t) \sim 1/t \)
  Nozieres, DeDominicis 1969 \( \rightarrow \) **not applicable** here!

- For the singular integral equation
  find a new integral operator \( K_1 \) such that
  is non-singular. Mushkelishvili 1953

- Method can be extended to Non-Abelian and 3D generalizations of the Kitaev model.
Remains exactly solvable with fluxes replaced by flux lines.

Spectrum has a gapless line. Additional three spin interaction induces Weyl Spin Liquid.

Mandal et al. 2009, Lee et al. 2014/15; Kimchi 2014

Hermanns et al. 2015
New features from inequivalent bonds and multiple bands → no orthogonality catastrophe

Distinct (high-energy) features of the Majorana fermions in powder averaged structure factor!
→ relation to $\beta$-Li$_2$IrO$_3$?
Results isotropic 3D QSL

- Single crystal dynamical structure factor along a path through the BZ

3. Exact dynamical correlation functions
Raman Scattering

- **Photon-in photon-out process** Devereaux, Hackl 2007
  - Polarization dependent

- **Polarization independence in QSLs** Cepas, Lhuillier 2008; Ko et al. 2010

- **Loudon-Fleury correlation function** Shastry, Shraiman 1990

\[
I(\omega) = \int_{-\infty}^{\infty} dt e^{i\omega t} \langle \hat{R}(t) \hat{R}(0) \rangle
\]

- **Na$_2$IrO$_3$ described by Kitaev-Heisenberg model** Jackeli, Khaliullin 2009

\[
\hat{H} = -J_K \sum_{\langle ij \rangle_a} \hat{\sigma}_i^a \hat{\sigma}_j^a + J_H \sum_{\langle ij \rangle} \hat{\sigma}_i \cdot \hat{\sigma}_j
\]

- **Vertex:**

\[
\hat{R} = \sum_{\langle ij \rangle_a} (\hat{\epsilon}_{\text{in}} \cdot \hat{d}_a)(\hat{\epsilon}_{\text{out}} \cdot \hat{d}_a) \left( K_K \hat{\sigma}_i^a \hat{\sigma}_j^a + K_H \hat{\sigma}_i \cdot \hat{\sigma}_j \right)
\]
3. Exact dynamical correlation functions

Raman Scattering Theory

- Close to the integrable limit
  \[ \frac{\lambda}{\lambda} = \frac{J_H}{J_K} = \frac{K_H}{K_K} \ll 1 \]

\[ I(\omega) \approx I_K(\omega) + I_H(\omega) \]

- Exactly solvable contribution of the integrable Kitaev model:

\[ I_K(\omega) = \int_{-\infty}^{\infty} dt e^{i\omega t} \langle 0 | T[\hat{R}_K(t)\hat{R}_K(0)] | 0 \rangle \]

\[ I_K(\omega) = 4\pi \sum_q \delta(\omega - 4|S(q)|) \left[ \frac{\text{Im} [h(q)S^*(q)]}{|S(q)|} \right]^2 \]

with

\[ h(q) = \sum_{\alpha} K_K (\hat{e}_{\text{in}} \cdot n_\alpha) (\hat{e}_{\text{out}} \cdot n_\alpha) e^{iqn_\alpha} \]

- Polarization independent!
Raman Scattering Theory

- Integrability-breaking Heisenberg term:

\[
I_H(\omega) = \int_{-\infty}^{\infty} dt e^{i\omega t} \langle 0 | T[\hat{R}_H(t)\hat{R}_H(0)]|0\rangle
\]

- Selection rule from flux sector
  
  e.g. for

\[
\langle \sigma_{Ar} y(t)\sigma_{Br} y(t)\sigma_{Ar} y(0)\sigma_{Br} y(0) \rangle
\]

- Quantum quench of 4 fluxes:

\[
I_H(\omega) = \int_{-\infty}^{\infty} dt e^{i\omega t} \sum_{a=x,y,z} \sum_{r} \langle M_0 | e^{it\hat{H}_0} e^{-it(\hat{H}_0 + \hat{V}_r)} | M_0 \rangle
\]

- Sharp contribution at the flux gap from g.s. contribution

\[
I_H^{[0]}(\omega) = 2\pi \delta (\omega - \Delta_F) \sqrt{\det (\mathcal{X}^\dagger \mathcal{X})}
\]

3. Exact dynamical correlation functions
Raman Scattering Results

- Weak polarization dependence
- Sharp response at the four flux gap
- Big 'hump' with fine features of the Majorana DOS

Gupta, Singh et al. 2014

RuCl$_3$

(interesting T-dependence)
3D QSL Raman scattering

- Polarization dependence from inequivalent bonds. → distinguishable from phonons

- With time reversal breaking three spin interaction $\kappa$ the low energy two spinon-DOS is changed → signatures of the Weyl points
4. Conclusion

- Topologically ordered phases beyond the Landau paradigm:
  - QSLs are one example with recent 2D/3D candidate materials.
5. Conclusion

- Exact dynamical correlation functions of a 2D/3D quantum spin liquid
  → Benchmark for numerics and approximations
  → Connections to experiments e.g. Li/Na$_2$IrO$_3$ and RuCl$_3$

- Fractionalized d.o.f. show up as characteristic features in the response
  → Flux gap + δ-peaks
  → Majorana fermion DOS

- Non-equilibrium physics beyond the X-ray edge

- INS, ESR and Raman experiments can diagnose spin liquids!
Thank you!

featured with a Viewpoint in Physics by A. Tsvelik


arXiv:1504.08037
arXiv:1507.01639 (accepted in PRB)
arXiv:1507.02865 (accepted in PRB)
arXiv:1508.05324

more coming soon…