Dynamics of a two-dimensional quantum spin-orbital liquid: spectroscopic signatures of fermionic magnons

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We provide an exact study of dynamical correlations for the quantum spin-orbital liquid phases of an SU(2)-symmetric Kitaev honeycomb lattice model. We show that the spin dynamics in this Kugel-Khomskii type model is exactly the density-density correlation function of $S = 1$ fermionic magnons, which could be probed in resonant inelastic x-ray scattering experiments. We also predict the characteristic signatures of spin-orbital fractionalization in inelastic neutron scattering experiments and compare them to the ones of the spin-anisotropic Kitaev honeycomb spin liquid.

Phases of matter which remain disordered down to lowest temperatures because of quantum fluctuations have fascinated the condensed matter community for a long time. One reason for this enduring interest is that they can host long-range entangled ground states displaying topological order \cite{1}. Quantum spin liquids (QSLs) \cite{2, 3} are prominent examples of such phases that have not been conclusively identified in experiment, in spite of the availability of many candidate materials. In addition to the absence of local order, the main reason for this long ongoing search for a QSL is the unusual nature of its excitations which carry only fractions of the usual quantum numbers probed experimentally. For example, an $S = 1$ spin flip excitation, diagnosed via the dynamical structure factor (DSF) in inelastic neutron scattering (INS), decays into multiple excitations, e.g. spinons and visons \cite{4} or Majorana fermions and fluxes \cite{5}, leading only to a broad featureless continuum response. An additional obstacle in this ongoing search is the fact that quantum liquids are inherently strongly interacting which makes it difficult to obtain rigorous theoretical predictions that could be compared to experiments beyond one-dimensional model cases.

An important conceptual development was the advent of exactly soluble models with QSL phases. The most prominent is the Kitaev honeycomb lattice model \cite{6–8}, which has permitted the calculation of exact results for dynamical correlations in the thermodynamic limit as probed in scattering experiments \cite{5, 9, 10}. The fractionalized excitations of the Kitaev spin liquid (KSL) are Majorana fermions in a plaquette flux background. The prediction that the Kitaev model could be relevant to specific heavy-ion Mott insulators (the Kitaev materials) \cite{11} rapidly followed by their synthesis \cite{7, 12, 13} provided additional motivation to evaluate dynamical response functions of a variety of scattering experiments \cite{5, 10, 14–24}. Unfortunately, most Kitaev materials \cite{7, 12, 13} show residual long range magnetic order instead of a pristine KSL phase, an observation well-explained by more complete models beyond the pure Kitaev limit \cite{25–27}. Nevertheless, the main features of the INS response of the Kitaev candidate material $\alpha$-RuCl$_3$ is arguably captured by the DSF of the Kitaev model \cite{28–32}. In spite of these recent developments, our understanding – even of the basic phenomenology and experimental signatures – of quantum liquids beyond the pure Kitaev model remains limited.

Here, we provide exact results for the dynamical response of a quantum spin-orbital liquid (QSOL) as found in certain Kugel-Khomskii (KK) models \cite{33–37}. We focus on systems with four degrees of freedom per site which are either equivalent to $j = 3/2$ spin models or KK models with doubly degenerate orbitals \cite{38, 39}. Thereby, we uncover qualitative differences to QSLs of the anisotropic $j = 1/2$ Kitaev type. In particular, we show that in a QSOL a $S = 1$ spin flip can excite only one type of excitation, e.g. two Majorana fermions without additional fluxes, leading to a much cleaner signature of fractionalization with a distinct momentum dependence absent in the KSL.

We compute the dynamical correlation functions of the SU(2)-symmetric Kitaev model

$$H = -\sum_{\langle lm \rangle \gamma} J_\gamma \left(T_\gamma^l \sigma_l \right) \cdot \left(T_\gamma^m \sigma_m \right),$$

which is a generalization of the spin anisotropic Kitaev model \cite{37}. Here, $J_\gamma$ are bond-dependent exchange constants, $T$ and $\sigma$ are orbital and spin operators satisfying $[T_\alpha, T_\beta] = 2\delta_{\alpha\beta}e^{\alpha\beta\gamma}T_\gamma$, $[\sigma_\alpha, \sigma_\beta] = 2\delta_{\alpha\beta}e^{\alpha\beta\gamma}\sigma_\gamma$ and $[T_\alpha, \sigma_\beta] = 0$. The model is another rare example of an exactly soluble one using a Majorana fermion representation of the spin-orbital operators $\sigma_\gamma$. It displays a QSOL ground state with an emergent $Z_2$ gauge field and fermionic excitations of the Majorana type related to spin flips dubbed fermionic magnons \cite{37}.

We find that the dynamical response of spin operators is given by the the density-density correlation $I_\gamma(q, \omega)$ of fermionic excitations, which can be probed with resonant inelastic x-ray scattering (RIXS) if $H$ is regarded as a $j = 3/2$ model \cite{41}. The DSF of the model is a linear combination of the flux diagonal part $I_\gamma(q, \omega)$ and a correlation function among the operators $\sigma^\alpha T^\beta$ exciting
both types of excitations.

The model - The SU(2)-symmetric Kitaev model has a macroscopic set of conserved plaquette operators $W_p$ analogous to the ones in the spin-1/2 Kitaev model \[6, 37\]. A key difference is that each $W_p$ affects only the orbital degrees of freedom of Eq. (1) and trivially commutes with all spin operators. The ground state of $H$ is easily found in an enlarged Hilbert space defined by a six-flavor Majorana representation of $\sigma$ and $T$: $\sigma^\alpha_i = -\frac{i}{2} e^{i \beta_i} \eta_i^\alpha \eta_i^\beta$ and $T^\alpha = -\frac{i}{2} e^{i \beta_i} \bar{\eta}_i^\alpha \bar{\eta}_i^\beta$ \[37, 40\]. The physical states are eigenstates of the projector $D_i = i \eta_i^\alpha \eta_i^\beta \bar{\eta}_i^\alpha \bar{\eta}_i^\beta$ with eigenvalue +1. This constraint also entails that $\sigma^\alpha_i T^\beta_i = -i \eta_i^\beta \bar{\eta}_i^\beta$ and allows to represent Eq. (1) like \[37\]

$$H = \sum_{\langle ij \rangle} \sum_\alpha J_{\langle ij \rangle, \alpha} \hat{u}_{\langle ij \rangle, \alpha} \eta_i^\alpha \eta_j^\alpha,$$

(2)

where $\hat{u}_{\langle ij \rangle, \alpha} = \eta_i^\alpha \eta_j^\beta$ is a $\mathbb{Z}_2$ gauge operator defined along the bond $\langle ij \rangle$ with $i$ on the even sublattice.

Note, Eq. (2) generalizes the fermionic representation of the spin-1/2 Kitaev model \[6\] by the presence of three Majorana flavors instead of one. Any eigenstate $|\psi\rangle$ of $H$ is then a direct product $|\psi\rangle = |F_\psi\rangle \otimes \prod_\alpha |M_\psi^\alpha\rangle \equiv |F_\psi\rangle \otimes |M_\psi\rangle$, where $|F_\psi\rangle$ is the flux sector and $|M_\psi^\alpha\rangle$ is a state for the $\eta^\alpha$ Majorana flavor of the “matter” sector. Lieb’s theorem \[42\] asserts that the global ground state is found in the flux sector $|F_0\rangle$ characterized by $W_p |F_0\rangle = |F_0\rangle$ for all plaquettes. In the language of $\mathbb{Z}_2$ gauge operators, $|F_0\rangle$ is obtained after fixing $u_{\langle ij \rangle, \alpha} = 1$ for all gauge fields. The translational symmetry of the ground state Hamiltonian $H_0$ permits a diagonalization in reciprocal space

$$H_0 = \sum_\alpha \sum_\alpha |\mu_\alpha\rangle (2 \sigma^\alpha \sigma^\alpha - 1)$$

(3)

where $\mu_\alpha = \sum_\gamma J_\gamma \exp(iq \cdot n_\gamma)$ with $n_{x,y} = (\pm \frac{1}{2}, \sqrt{x})$ and $n_z = 0$. The excitations in the matter sector have a momentum-dependent dispersion $\epsilon_\alpha = 2 |\mu_\alpha|$ that can be gapless or a gapped according to the values of $J_\gamma$ \[6\].

It is instructive to analyze the fractionalization processes implied by Eq. (2). We recall that the spin fractionalization in the standard Kitaev model can be represented by $\sigma \sim e^{i \epsilon_m c}$, where $\epsilon$ and $m$ are visons corresponding to the insertion of $\pi$-fluxes in two adjacent plaquettes and $c$ is the Majorana fermion \[3, 6, 9\]. The same kind of fractionalization occurs here but now for the spin-orbital operators like $\sigma^\alpha T^\beta \sim e^{i \epsilon_m c}$. In this case, the $\epsilon$ and $m$ particles only affect the orbital sector and the three $\epsilon$ particles correspond to the Majorana flavors $\eta^\alpha$ for spins. As a qualitatively new feature of the QSOL, the spin $\sigma^\alpha$ fractionalizes into two $\epsilon$ particles unrelated to the formation of visons which we show in the following translates into qualitatively distinct features in the dynamical response.

Dynamical correlation functions - We treat the SU(2)-symmetric Kitaev model as a model of $j = 3/2$ effective moments realized in 4D Mott insulators, since this allows us to associate the dynamical correlations to the expected responses of both RXXS and INS \[41\]. Our goal is to compute the DSF given by the Fourier transform of the correlation function $S_{\alpha\beta}(q,t) = \langle \psi_0 | \hat{J}_{\alpha\beta}^z(t) \hat{J}_{\alpha\beta}^z(0) | \psi_0 \rangle$, of the angular momentum operators \[41\]

$$j^\alpha_i \equiv \frac{1}{2} \sigma^\alpha_i - \sigma^\alpha_i T^{(\alpha)}_i$$

(4)

where $\alpha = x,y,z$, $T^i_\gamma(z) = T^i_\gamma$ and $T^i_\gamma(x,y) = -\frac{1}{2} T^z_\gamma \pm \frac{\sqrt{3}}{2} T^y_\gamma$.

It turns out that the DSF is only a sum of two contributions because $\langle \sigma^\alpha_i(t) \sigma^\alpha_i T^{(\alpha)}_i(0) \rangle = \langle \sigma^\alpha_i T^z_i(t) \sigma^\alpha_i(0) \rangle = 0$, since the action of $\sigma^\alpha_i T^{(\alpha)}_i$ on $|\psi_0\rangle$ involves the creation of a pair of vison whereas $\sigma^\alpha_i T^{(\alpha)}_i$ is flux-conserving.

First, we discuss the correlation function $I_{ij}^{(x)}(t) = \langle \sigma^\alpha_i(t) \sigma^\alpha_j(0) \rangle$. The application of $\sigma^\beta_i$ on $|\psi_0\rangle$ preserves the gauge fluxes, thus allowing the evaluation of $I_{ij}^{(x)}(t)$ in terms of ground state correlations of Majorana fermions \[39\]. Additionally, since the Hamiltonian is diagonal in the Majorana flavor index we find $I_{ij}^{(x)}(t) \propto \delta_{ij}$ and the SU(2) symmetry implies that $I_{ij}^{(x)}(t)$ is isotropic for all $\alpha = x,y,z$. Hence, we only need to evaluate a single (we omitted the $zz$-label)

$$I_{im}(t) = -\sum_\lambda e^{i(E_0 - E_\lambda)t} \langle M_0 | \eta^\lambda_\eta^\mu_\lambda | \lambda \rangle \langle \lambda | \eta^\lambda_\eta^\mu_\lambda | M_0 \rangle$$

(5)

where the sum runs over all two-particle excitations of $|M_0\rangle$ and $|M_0^\lambda\rangle$. A convenient representation of Eq. (5) is given in terms of $S = 1$ fermionic magnons defined by $c^\dagger_\lambda = \frac{1}{2} (\eta^\lambda + i \eta^\mu_\lambda)$ \[37\]. After performing the Fourier transform, the spin-spin correlation reads

$$I(q, \omega) = \frac{8\pi}{N} \sum_{R_l, R_m} e^{iq \cdot (R_m - R_l)} \delta(\omega - (E_\lambda - E_0)) \times \langle M_0 | n^\lambda_{R_l} | \lambda \rangle \langle \lambda | n^\lambda_{R_m} | M_0 \rangle$$

$$\equiv \frac{2\pi}{N} \sum_{k} \delta(\omega - 2 (\mu_k + \mu_{k+q})) \times |1 - e^{2i (\theta_k + \theta_{k+q})}|^2$$

(6)

where $n^\lambda_{R_l}$ is the total number of $c$ fermions at the $R_l$ unit cell and $e^{-2i \theta_k} = \mu_k / |\mu_k|$. The first equation shows that $I(q, \omega)$ is readily interpreted as the density-density correlations of fermionic magnons. In contrast to the
KSLs, the real-space spin–spin correlations decay algebraically (exponentially) for gapless (gapped) fermionic dispersion [37]. Remarkably, the splitting between spin and orbital degrees of freedom allowed a simple, yet exact, expression for the dynamics of a QSOL with longer range correlations.

Second, we show how the correlation functions \( W_{lm}^{\alpha \beta, \delta \gamma}(t) \) are mapped into a quantum quench problem similar to the one discussed for KSLs. Following the arguments of Ref. [9], \( W_{lm}^{\alpha \beta, \delta \gamma} = \left\langle \sigma_i^{\alpha} T_{i}^{\delta} (t) \sigma_m^{\beta} T_{m}^{\gamma} (0) \right\rangle \) will be non-zero only if \( l = m \) or if \( l \) and \( m \) are nearest neighbors. Fixing \( l(m) \) to the even (odd) sublattice, \( W_{lm}^{\alpha \beta, \delta \gamma}(t) \) is obtained from

\[
\left\langle \hat{\sigma}_i^{\alpha} T_{i}^{\delta} (t) \hat{\sigma}_m^{\beta} T_{m}^{\gamma} (0) \right\rangle = -i \left\langle M_0 \eta_i^{\alpha} e^{-i(H_0+V_\gamma) t} \eta_m^{\beta} \right\rangle \times e^{i E_0 t} \delta_i^{\alpha} \delta_m^{\beta} \delta_{r(m),r(l)+n},
\]

in which \( E_0 \) is the ground state energy, \( r(l) \) is the unit cell containing the site \( l \), and

\[
V_\gamma = -2J_{\gamma} \sum_{\alpha} \left[ \eta_{\alpha,l}(l) \eta_{\alpha,r(l)+n}^{\dagger} \right] = \sum_{\alpha} V^{(\alpha)}_\gamma.
\]

This expression differs from the quantum quenches obtained in the spin-1/2 Kitaev model [5, 14, 15, 17, 18] by the number of flavors in \( H_0, V_\gamma \) and \( |M_0\rangle \). Therefore, the non-zero matrix elements have the form

\[
W_{lm, \gamma}^{\alpha \alpha}(t) = W_{lm, \gamma}^{\alpha \alpha}(t) \prod_{\delta \neq \alpha} L_{\gamma}^{\delta}(t) \delta_{r(m),r(l)+n},
\]

where

\[
W_{lm, \gamma}^{\alpha \alpha}(t) = -i e^{i E_0 t} \left\langle M_0^{\alpha} \eta_i^{\alpha} e^{-i(H_0^{(\alpha)}+V_\gamma^{(\alpha)}) t} \eta_m^{\alpha} \right\rangle |M_0^{\alpha}\rangle.
\]

The matrix element in \( W_{lm, \gamma}^{\alpha \alpha}(t) \) is the same of the Kitaev model [5, 14, 17] but the multiple flavors matter result in a new time-dependent phase \( L_{\gamma}^{\delta}(t) \) which can be calculated exactly via a Pfaffian formula from functional integrals [14]. Finally, we exploit the SU(2) invariance of the model implying that \( W_{lm, \gamma}^{\alpha \alpha} \) is flavor independent and focus on \( \alpha = z \).

Overall, the DSF of the SU(2)-symmetric model is given by

\[
S(\mathbf{q}, \omega) = \frac{3}{4} I(\mathbf{q}, \omega) + \frac{3}{2} (W_2 + W_3)(\mathbf{q}, \omega),
\]

where \( W_\gamma(\mathbf{q}, \omega) \) is the Fourier transform of \( W_{lm, \gamma}(t) \). Notice that correlations along the \( y \)-bonds do not contribute to the DSF as predicted by the absence of the operators \( T_t^y \) in Eq. (4). Physically, this reflects the absence of coupling between the neutron spin and the \( \sigma_i^{\alpha} T_{i}^{\beta} \) operators due to their evenness under time-reversal [41, 43].

**Results** - Let us first discuss the density-density correlation of fermionic magnons \( I(\mathbf{q}, \omega) \) from dynamical spin correlations presented in Eq. (6). Note, for our choice of orbital representation it is directly measurable with RIXS at the L_3-edge [41]. The responses displayed in Fig. 1 strongly depend upon the value of the transferred momenta \( \mathbf{q} \) in contrast to the DSF of the spin-1/2 Kitaev model whose ultra short ranged spin correlations result in an almost dispersionless response [5, 14, 17].

An analysis of \( I(\mathbf{q} = \mathbf{K}, \omega) \) shows that they closely follow the density of states \( \rho(\omega) \) of two-fermion excitations (lower panel), e.g. with intensity peaks related to the van Hove singularities of the fermionic bands. In contrast to the gapped response of the spin-1/2 Kitaev model even for gapless fermions, one would expect a verifiable response of \( I(\mathbf{q}, \omega) \) for excitations below the vison gap in gapless QSOs because of the different flux selection rules, especially when \( \mathbf{q} \approx \Gamma \). However, the form factor of \( I(\mathbf{q}, \omega) \) vanishes at \( \mathbf{q} = \Gamma \) which results in zero intensity at this point. This feature can be explained...
via the form factor at \( q = \Gamma \) which is proportional to \(|\langle \hat{M}_0 \rangle|^2\). Since \( |\hat{M}_0 \rangle \) must be a many-body singlet of \( \sigma, \sum_i \sigma_i^z |\hat{M}_0 \rangle = 0 \) and the response is zero [41].

It is interesting to note that the dynamical spin response \( I(q, \omega) \) of the QSOL is similar to the RIXS response of the spin-1/2 Kitaev model [20]. The form factor in both cases is proportional to the term \( |1 - e^{2i(\theta_{k+q} - \theta_k)}|^2 \), which is a direct consequence of the projective transformations of fermions under inversion [20]. However, in the case of the spin-1/2 Kitaev model, the form factor still arises from nearest neighbor correlations, which generates an additional factor \(|\langle \hat{M}_0 \rangle|^2\). Therefore, the response of the QSOL \( I(q, \omega) \) has a stronger intensity at lower energies and a more pronounced momentum dependence.

We now turn to the dynamical correlations of the spin-orbital operators displayed in Fig. 2(a-c). The response \((W_z + W_x)(q, \omega)\) is qualitatively similar to the DSF of the spin-1/2 Kitaev model [5, 14, 17]. There is a flux gap even in the gapless phase and only weak dependence on the transferred momentum. However, there are two important differences due to the additional Majorana flavors: the flux gap is three times larger and the response extends to energies beyond the Majorana fermion band width (shifted by the gap). These higher-energy excitations originate from the extra phases \( I_q^\alpha(t) \) in Eq. (9) and have a simple interpretation: the action of a spin-orbital operator excites one flavor of Majorana fermions and a pair of fluxes, the latter also shaking up the remaining two flavor sectors without fermion excitations resulting in the Loschmidt echo-like quench \( I_q^\alpha(t) \).

Finally, the sum of the contributions, see Eq. (10), is the DSF as measurable in INS shown in Fig. 2(d-f). The DSF displays mixed characteristics of the dynamics of fermionic magnons and the correlation of spin-orbital operators. Our exact results provide a concrete example of how RIXS can complement studies of INS to disentangle the different signatures of quantum number fractionalization related to the spin and orbital degrees of freedom in QSOLs. While RIXS measures the dispersion of fermionic excitations but not the flux gap, INS captures both features but is unable to distinguish them by itself.

**Experimental connections** - A natural question is: does our choice of exactly soluble QSOL capture the low energy physics of any candidate material? Ref. [37] proposed that a decorated honeycomb lattice can give rise to the SU(2)-symmetric Kitaev model but more promising seems to be the connection with spin-orbital systems. The bond-frustrated exchanges of Eq. (1) resembles those appearing in KK models [38, 44] associated with Mott insulators that retain \( e_g \) degeneracy [38, 44–47]. The synthesis of 4/5d\(^1\) Mott insulators with \( j = 3/2 \) magnetic moments [41, 48–52] or graphene-based superlattices [43, 53, 54] has recently increased the list of Kugel-Khomskii materials.

Interestingly, Eq. (1) is expected to emerge in highly anisotropic materials, e.g. coupled chains, because of the inherent difference between spin and orbital operators. While the spin transforms as \( \Theta \sigma \Theta^{-1} = -\sigma \) under time-reversal \( \Theta \) the orbital operators \( T_x \) and \( T_z \) are time-reversal invariant and \( \Theta T_y \Theta^{-1} = -T_y \) [41, 43]. This symmetry property implies that \( \sigma^a T^{x,y} \) must be a linear combination of dipoles and octupoles of an effective \( j = 3/2 \) angular moment while \( \sigma^a T^y \) are equivalent to quadrupoles of \( j \) [41]. The interactions along one of the bond directions is then of a different nature in solid-state implementations of Eq. (1). In general, the key ingredient of the model studied here is the SU(2) symmetry of spins which is common among several KK models with

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**Figure 2:** The correlation function \((W_z + W_x)(q, \omega)\) (upper) and the DSF as measured in INS of the SU(2)-symmetric Kitaev model for the cases (a+d) \( J_x = J_y = J_z \), (b+e) \( J_x = J_y, J_z = 0.7J_z \), and (c+f) \( J_x = J_y = 0.15J_x \).
possible QSOL ground states [41, 48–54]. Thus, the resulting rich phenomenology we have found for our exactly soluble QSOL is expected to be robust and provide a valuable guide for future experiments on candidate materials.

Conclusion - We provide the first exact results of dynamical correlations of a QSOL also giving an example for algebraically decaying spin liquids. Our computation of the dynamical spin- and orbital-correlations of an SU(2)-symmetric extension of the Kitaev model shows how spin-orbital fractionalization is manifest in scattering experiments like INS and RIXS. For example, it would be desirable to look for signatures of $S = 1$ fermionic magnons with a distinct energy and momentum dependence in Kugel-Khomskii materials with SU(2) symmetry.

In the future it would be desirable to extend the as of yet short list of rigorous results for the dynamics (and finite temperature properties [22–24]) of quantum liquids to other exactly soluble systems, e.g. SU(2)-symmetric Kitaev models on other tricoordinated lattices [15, 18, 21], models with a spinon Fermi sea [37, 55], or symmetric Kitaev models on other tricoordinated lattices [33–36, 56] whose dynamical correlations are also mapped to quantum quench problems.

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