Nontrivial Triplon Topology and Triplon Liquid in Kitaev-Heisenberg–type Excitonic Magnets

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The combination of strong spin-orbit coupling and correlations, e.g. in ruthenates and iridates, has been proposed as a means to realize quantum materials with nontrivial topological properties. We discuss here Mott insulators where onsite spin-orbit coupling favors a local \( J_{\text{tot}} = 0 \) singlet ground state. We investigate excitations into a low-lying triplet, triplons, and find them to acquire nontrivial band topology in a magnetic field. We also comment on magnetic states resulting from triplon condensation, where we find, in addition to the same ordered phases known from the \( J_{\text{tot}} = \frac{1}{2} \) Kitaev-Heisenberg model, a triplon liquid taking the parameter space of Kitaev's spin liquid.

Prime candidate systems for the interaction of spin-orbit coupling with substantial electronic correlations are those containing 4d and 5d transition metals, where 'topological Mott insulators' [1] or topological spin liquids were proposed. A prominent example is the prediction of Kitaev’s spin liquid [2] in materials with a single hole in the \( t_{2g} \) levels [3,4]. Strong research activity has subsequently focused on honeycomb iridates [5] and on \( \alpha-\text{RuCl}_3 \) [6,7]. Encouragingly, \( \text{H}_2\text{LiIr}_2\text{O}_6 \) does indeed not show magnetic order [8] and zig-zag order in \( \alpha-\text{RuCl}_3 \) can be suppressed by a magnetic field [9,10]. In the latter case, a thermal Hall effect due to the Majorana edge states has been reported [11].

Current interest has similarly been drawn to spin-orbit coupled Mott insulators with two holes in the \( t_{2g} \) shell. In addition to total spin \( S = 1 \), they would have an effective orbital angular momentum \( L = 1 \), and spin-orbit coupling prefers their opposite orientation into a singlet ground state \( J_{\text{tot}} = 0 \). On the other hand, magnetic superexchange between two ions involves the excited states with \( J_{\text{tot}} > 0 \). This superexchange can drive excitonic magnetism via the condensation of bosonic 'triplons' [12,13].

While the classical limit of this scenario is governed by the same symmetries – and thus by similar magnetic ordering patterns – as the \( J_{\text{tot}} = \frac{1}{2} \) scenario, the underlying degree of freedom is a superposition of the \( J_{\text{tot}} = 0 \) and \( J_{\text{tot}} = 1 \) states. In addition to opening the route to unconventional collective state like triplet superconductivity [14], this has a decisive impact on excitations, e.g. on their dispersion in the Brillouin zone. With the observation of an amplitude 'Higgs' mode, \( \text{Ca}_2\text{RuO}_4 \) has been argued to realize such a scenario close to a quantum critical point [15,16].

Here, we investigate this scenario on the honeycomb lattice, a model that should be appropriate to compounds like \( \text{Li}_2\text{RuO}_3 \) [17] and \( \text{Ag}_3\text{LiRu}_2\text{O}_6 \) [18], and whose low coordination number has been proposed to make it susceptible to states with enhanced quantum fluctuations [12]. We focus first on the regime with dominant \( J_{\text{tot}} = 0 \) character, i.e., where onsite spin-orbit coupling dominates over inter-site superexchange, as found for \( d^3 \) iridates with a double-perovskite lattice [19,20]. We find that excitations become topologically nontrivial in magnetic fields. This implies features like protected edge states crossing triplon-band gaps, similar to the topological magnon edge states discussed as spin conductors with reduced dissipation [22,23], and the thermal Hall effect [24–28].

We also present a phase diagram of the magnetic states emerging once the \( J_{\text{tot}} = 1 \) states become more dominant. We find magnetically ordered phases analogous to those of the \( J_{\text{tot}} = \frac{1}{2} \) Kitaev-Heisenberg model, and also a disordered phase taking the place of Kitaev’s spin liquid. This ‘triplon liquid’ realizes a quantum-mechanical order-by-disorder scenario, where quantum fluctuations select a unique gapped ground state from classically degenerate dimer coverings.

**Model.** Based on Ref. [12], we model the strongly spin-orbit coupled \( d^3 \) Mott insulators as

\[
H = \sum_{i,a} n_{i,a} + J \sum_{\langle i,j \rangle} \left( T_{i,j}^a T_{j,i}^a + H. c. \right) + K \sum_{\alpha} \sum_{\langle i,j \rangle} \left( T_{i,j,\alpha}^a T_{j,i,\alpha}^a - c_K T_{i,a}^\dagger T_{j,a}^\dagger + H. c. \right) + \Gamma \sum_{\alpha,\beta,\gamma} \sum_{\langle i,j \rangle} \left( T_{i,\alpha}^\dagger T_{j,\beta}^\dagger T_{j,\gamma}^\dagger T_{i,\gamma}^\dagger + H. c. \right),
\]

where \( T_{i,a}^\dagger \) (\( T_{i,a} \)) creates (annihilates) a triplon, i.e., a hard-core boson, with flavor \( \alpha = x,y,z \) at site \( i \). These operators are collected into vectors \( T_i = (T_{i,x}, T_{i,y}, T_{i,z}) \). The honeycomb lattice is built of three bond directions, here likewise labeled by \( \alpha = x,y,z \) so that the triplon with coupling \( K \) on a given bond bears the same index as the bond. Energy \( \lambda \) associated with creating a triplon is given by spin-orbit coupling separating the \( J_{\text{tot}} = 0 \) from the \( J_{\text{tot}} = 1 \) states. Couplings \( J, K, \) and \( \Gamma \) can be
FIG. 1. (a) Triplon bands for momentum $\mathbf{k}$ along the line $\Gamma = (0,0)$ to $K = (0, \frac{2\pi}{a})$ for dominant ‘Kitaev’ coupling, i.e. for $J = \Gamma = 0$, and deep within the $J_{\text{tot}} = 0$ regime, i.e. for $\lambda \gg K$. Bands are three-fold degenerate in the absence of a magnetic field and split for $h = h(1,1,1)$ with $h = 0.3K$. Inset indicates the first Brillouin zone with three high-symmetry points. (b) Topologically nontrivial bands, with Chern numbers $C = -1, 0, 1$ from the bottom to the top, and edge states along zig-zag edges, obtained for a cylinder. (c) Decorated honeycomb lattice realized for $J, \Gamma \approx 0$ in a magnetic field $h$ perpendicular to the plane. Thick colored lines are the bonds of the honeycomb lattice, triplons are confined to a bond for $h = 0$. Each shaded circle corresponds to one real-space site, $h = 0$ allows onsite flavor transitions illustrated via triangles. (d) Next-nearest-neighbor (NNN) Dzyaloshinskii-Moriya (DM) interactions $\lambda$. $D$ is positive (negative) for triplon hopping in the direction (opposite to) the arrows. (e) Examples for triplon configurations found in the triplon liquid; right- and left-facing triangles and circles stand for $x$-, $y$-, and $z$-type triplons.

estimated from second-order perturbation theory. The constants $c_J$, $c_K$, and $c_T$ giving the relative strength of triplon hopping to pair creation terms depend on the microscopic processes involved. However, they are of order 1, and since we have verified that the results presented here do not depend on their precise values, we set here $c_J = c_K = c_T = 1$. The full model also features three- and four-triplon terms, but as these only become relevant once the groundstate contains an appreciable number of triplons, their influence on triplon excitations of the $J_{\text{tot}} = 0$ state and on its ordering tendencies (before order sets it) are small, and they are neglected here.

For 90° bond angles, dominant oxygen-mediated electron hopping $t$ and neglecting Hund’s rule, $K$ becomes $\approx -J$ so that every triplon flavor can move on two kinds of bonds along a zig-zag line through the honeycomb lattice [12]. Hopping $t'$ due to direct overlap between the $d$ orbitals leads to $K \gg J > 0$; and if both $t$ and $t'$ are present, $\Gamma \approx t' \approx t$ becomes active. Further, Hund’s rule coupling promotes FM exchange [13], processes via $e_g$ orbitals might also contribute [4,29], and a honeycomb lattice can also arise with 180° bond angles in ‘dice-lattice’ bilayer heterostructures [30]. Since a large variety of parameter combinations are possible, we treat $J$, $K$ and $\Gamma$ as material-dependent and vary them in the present study.

Nontrivial triplon topology. For $\lambda \gg J, K, \Gamma$, the $J_{\text{tot}} = 0$ state determines the ground state, but once a triplon is excited, it can move to another site via the $T_i^T T_j^T$ terms of (1). The $T_i^T T_j^T$ terms enter in order $\frac{1}{\sqrt{N}}$, and we consequently neglect them in this analysis of excitations deep within the $J_{\text{tot}} = 0$ phase, see also Ref. [21]

The bands described by (1) have Chern number $C = 0$, but can nevertheless show edge states. These can be most easily seen for the extreme “Kitaev” limit $\lambda \gg |K| > 0$ and $J = \Gamma = 0$, where one finds two groups of threefold degenerate dispersionless bands at energies $\lambda \pm K$, see Fig. 1(a). Each corresponds to one triplon flavor and eigenstates are perfectly localized on isolated bonds of the honeycomb lattice, see Fig. 1(c). If a zig-zag edge cuts all $z$ bonds along a vertical line, $z$-triplon states on the edge sites have no site to hop to, so that their energy becomes $\lambda$ instead of $\pm K$, see Fig. 1(b). Such states can be ascribed a topological origin [31] that is related to the Zak phase [32] and to the topological end states of a Su-Shrieffer-Heeger (SSH) chain [33]. Very recently, a model supporting such states has been argued to describe neutron-scattering data for Ba$_2$CuSi$_2$O$_6$Cl$_2$ [34].

The ‘SSH’ edge states discussed above do not cross the gap between triplon bands, are localized to isolated sites for $|K| > |J||\Gamma|$, and would thus not be good candidates for transport. Edge states between bands with different Chern numbers, which do cross gaps and support a thermal Hall effect, need broken time-reversal symmetry. One possibility is a magnetic field $H_m = h \sum_i M_i$, which couples to the magnetic moment on site $i$,

$$M_i = -i\sqrt{6}(\mathbf{T}_i - \mathbf{T}_i^T) + ig\mathbf{T}_i^T \times \mathbf{T}_i^T,$$

with $g = \frac{1}{2}$ [12]. Again, the first term linear in triplon operators is suppressed at large $\lambda$.

The second term in (2), which drives onsite flavor transitions, can as before be discussed most clearly for the extreme “Kitaev” limit $\lambda \gg |K| > 0$ and $J = \Gamma = 0$. Starting from the degenerate dispersionless bands of Fig. 1(a), a field $h \parallel (1,1,1)$ [i.e. perpendicular to the honeycomb plane] allows transitions between flavors on each site. Triplons are then no longer localized to a single bond and bands become dispersive, see Fig. 1(a) and (b). As illustrated in the cartoon Fig. 1(c), the system in fact becomes equivalent to a decorated honeycomb lattice, where topologically nontrivial bands can arise [33]. As a result of the imaginary phase $i$, see Eq. (2) and Fig. 1(c), the top and bottom band of each triplet acquires a non-trivial Chern number $C = \pm 1$, and the two bands are connected by protected edge states, see Fig. 1(b).

Figure 2 gives a phase diagram in $J-K$ parameter space, with topologically nontrivial bands almost everywhere. Gaps between Chern bands can be quite small.
and energy ranges of bands may in fact overlap with indirect gaps; more robust gaps are generally found for intermediate $\alpha$ (i.e. for large $K$). Allowing $\Gamma \neq 0$ significantly affects phase boundaries (not shown), but topological band character persists. In general, finite magnetic fields are needed, but correspond to achievable strengths of a few Tesla for estimated parameters [12]. This implies that $t^{3}_{1g}$ honeycomb insulators provide a viable route to the observation of triplon bands with Chern numbers as high as $C = 5$.

Nontrivial triplon topology in coupled intersite-dimer systems arises through DM interactions [24, 25, 31, 35], which are symmetry-allowed on NNN-bonds and take the form:

$$H_{DM} = \sum_{\langle i,j \rangle} D_{ij} \cdot T^{\dagger}_{i} \times T_{j},$$

(3)

with $D_{ij} = \pm D(1, 1, 1)$, i.e. perpendicular to the plane; $\langle i,j \rangle$ denotes NNNs and the + (-) sign applies to (anti-)clockwise motion within a hexagon, see Fig. 1(d). The similarity of DM term [33] and magnetization [2] is obvious. We have found DM interactions to support Chern numbers $C = \pm 1$ in the absence of a magnetic field, e.g. for $J = 1$, $\Gamma = K = 0$, and $h = 0$. However, the gaps are here rather fragile and nontrivial band topology is lost for finite $K$ and $\Gamma$ of the order of $D$. As NNN DM terms are in general expected to be rather smaller than NN interactions $K$ and $\Gamma$, this suggests a minor role for the former [56].

**Magnetic Phase Diagram and triplon liquid.** While a detailed investigation of the model's magnetic phases is beyond the scope of this work [37], we shortly discuss their basic features. The ordering vector expected for a magnetically ordered phase is the one where the triplon excitations first reach zero energy. The $T^{\dagger}_{i}T_{j}$ terms in the Hamiltonian have to be included here. We have accordingly used a Bogoliubov-de Gennes transformation [38], which neglects the hard-core constraint of the triplons, and exact diagonalization (ED), which is restricted to small clusters. We additionally interpolated between these two approaches using cluster-perturbation theory, which incorporates the hard-core constraint within the directly solved cluster.

For $\Gamma = 0$, the phase diagram obtained from ED is given in Fig. 3(a). The dark region in the middle corresponds to the $J_{tot} = 0$ regime, where hardly any triplons are mixed into the ground state $|\phi_{0}\rangle$ and where magnetic structure factors

$$S^{\alpha,\beta}(k) = \| \sum_{i} e^{i k \cdot r_{i}} (T^{\dagger}_{i,\alpha} - T_{i,\beta}) |\phi_{0}\rangle \|^2$$

(4)

are thus small for any $k$. Ground-state fidelity in
Fig. 3(b) as well as the second derivative of the ground-state energy in Fig. 3(c) have here a single peak, which indicates a first-order phase transition. The canonical-boson treatment and cluster-perturbation theory agree with these phase boundaries, which furthermore correspond more closely to classical predictions than in the $J_{\text{tot}} = \frac{1}{2}$ Kitaev-Heisenberg model. As in a classical model, our phase diagram going from 0 to 180° (i.e. for $K > 0$) perfectly repeats itself for the negative-$K$ part going from 180° to 360° (except that FM and AF change places and that zigzag becomes stripy).

Differences between the classical analysis and ED arise near the the ‘Kitaev’ limits $J = \Gamma = 0$. Fidelity and second energy derivative obtained from ED, see Figs. 3(b,c), argue here against the single first-order transition of the classical scenario and in favor of an intermediate phase in a narrow but finite parameter regime around the Kitaev points. We have found the phase to be stable against small $\Gamma \neq 0$, the stability range of this intermediate ‘triplon liquid’ is again not unlike that of Kitaev’s spin liquid in the corresponding $J_{\text{tot}} = \frac{1}{2}$ model.

For $J = \Gamma = 0$, the ground state contains, in addition to the vacuum state, only configurations where $x$- ($y$-, $z$-) bosons sit on both ends of $x$- ($y$-, $z$-) bonds, see Fig. 1(e) for examples. This observation allows us to restrict the Hilbert space to such dimer configurations and to obtain ground states of clusters with up to 30 sites; excitations going beyond this restricted Hilbert space can be obtained for up to 18 sites. Based on the dimer observation, one can moreover see that any structure factors are strictly short range, and we find numerically no indications for bond order either. With a triplon density $n = 0.33$ at large $K$ (somewhat below the $n = 0.45$ of the ordered phases), the phase clearly differs from the vacuum with $n = 0$, and we term it a ‘triplon liquid’.

Semi-classically, we expect for $J = \Gamma = 0$ an infinitely degenerate ground-state manifold of dimer coverings, with each dimer in a superposition of ‘empty’ and ‘occupied’ and an energy of $E_c = -K/2$ per site for $K \gg \lambda$. The triplon liquid has a non-degenerate ground state with markedly lower energy. This quantum-mechanical order-by-disorder mechanism is largely mediated by the vacuum state, which is shared between the dimer coverings by-disorder mechanism is largely mediated by the vacuum state, as for materials like $\text{Li}_2\text{RuO}_3$ [17] and $\text{Ag}_2\text{LiRu}_2\text{O}_6$ [18].

For strong intersite superexchange, we find magnetically ordered states (Néel, stripe and zig-zag AF and FM) as well as a triplon liquid stabilized out of classically degenerate dimer coverings via a quantum order-by-disorder mechanism.

At weaker superexchange, where the ground state is dominated by the $J_{\text{tot}} = 0$ state of the ion, excitations are found to be topologically nontrivial as soon as orbital anisotropies become relevant. Topologically nontrivial triplon bands have been proposed [24] and found to agree with neutron scattering data for $\text{SrCu}_2(\text{BO}_3)_2$, whose ground state consists of singlets on dimers; the discussion has since been extended to other geometries [25, 31, 35]. Topological triplon states in these dimer systems rely on DM interactions, which we found to compete with symmetric anisotropic exchange in the present $\text{ onsite}$-singlet systems. Consequently, magnetic fields perpendicular to the plane appear a more promising route towards nontrivial triplon topology when anisotropic couplings can be expected to dominate over DM interactions.

In addition to the $J_{\text{tot}} = 0$ regime discussed above, topologically nontrivial excitations are expected to persist into the FM phase, analogous to the nontrivial magnon topology in ferromagnetically polarized states of the $J_{\text{tot}} = \frac{1}{2}$ Kitaev model [22, 33]. The AF patterns require a more detailed symmetry analysis [34], but may also harbor nontrivial magnon bands. Finally, potential topological properties of the triplon liquid present an intriguing question once a magnetic field renders the underlying single-triplon states nontrivial.

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