Twisted superfluid and supersolid phases of triplons in bilayer honeycomb magnets

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We demonstrate that low-lying triplon excitations in a bilayer Heisenberg antiferromagnet provide a promising avenue to realise magnetic analogs of twisted superfluid and supersolid phases that were recently reported for two-component ultracold atomic condensate in an optical lattice. Using a cluster Gutzwiller mean field theory, we establish that Dzyaloshinskii-Moriya interactions (DMI), that are common in many quantum magnets, stabilize these phases in magnetic system, in contrast to pair hopping process that is necessary for ultracold atoms. The critical value of DMI for transition to the twisted superfluid and twisted supersolid phases depends on the strength of the (frustrated) interlayer interactions that can be tuned by applying external pressure on and / or shearing force between the layers. Furthermore, we show that the strength of DMI can be controllably varied by coupling to tailored circularly polarized light. Our results provide crucial guidance for the experimental search of twisted superfluid and supersolid phases of triplons in real quantum magnets.

I. INTRODUCTION

The observation of twisted, multi-orbital superfluid in binary mixtures of ultracold $^{87}\text{Rb}$ atoms in two different hyperfine states on a honeycomb optical lattice has attracted heightened interest in this novel quantum state of matter [1]. The twisted superfluid (or twisted supersolid) state is characterised by a complex order parameter – the phase of the local superfluid order parameter at each site changes continuously forming a “twisting pattern”, thus breaking time reversal symmetry spontaneously. Interestingly, complex order parameters have experimentally been shown to be associated with other novel strongly correlated phases such as the superconducting states of Sr$_2$RuO$_4$ [2, 3] and UPt$_3$ [4] and the pseudo-gap state in the cuprate high-T$_c$ superconductor, B-2212 [5, 6]. A detailed understanding of the twisted superfluid state can help gain insight into these states as well. Subsequent theoretical studies have shown that the extended Bose Hubbard model with an additional pair hopping term can stabilize a twisted superfluid (TSF) ground state over a finite range of parameters [7].

Quantum magnets have long served as a versatile platform for realizing magnonic analogs of complex bosonic phases, often under less extreme conditions. For example, temperature needed for Bose Einstein condensation (BEC) of magnons varies from a few Kelvins in many quantum magnets [8–10] to room temperature in Yttrium Iron Garnet (YIG) thin films [11, 12], in contrast to nano-Kelvin temperature scales required for BEC in ultracold atoms [13, 14]. In this work, we show that twisted superfluid and twisted supersolid phases of magnons are realized in a bilayer honeycomb Heisenberg model. Interestingly, pair hopping process of magnons is not essential for stabilizing these phases [7], in contrast to ultracold atomic systems. Instead, a next nearest neighbor (NNN) Dzyaloshinskii-Moriya interaction (DMI) – which is present in many quantum magnets – is sufficient to yield field induced twisted superfluid (TSF) and twisted supersolid (TSS) phases over wide ranges of parameters.

Our paper is structured as follows. In Sec. II, we introduce the microscopic spin Hamiltonian and describe the physics in the non-interacting limit by deriving the tight-binding triplon Hamiltonian and triplon band structure. The cluster Gutzwiller mean field theory (CGMFT) is introduced in Sec. III which is used to calculate order-parameters in interacting limit. In Sec. IV we present the results of our study in the form of the order parameters and phase diagram as a function of magnetic field, DMI and Heisenberg interactions. In Sec.V, we propose the possible materials in search of TSS and TSF phases. Finally, in Sec. V, we show that circularly polarized light can be used to induce DMI greater than the cutoff DMI required to realize TSS (TSF), whereas presence of frustration among interlayer and intralayer interactions is helpful to lower the cutoff value of DMI (see also Appendix. C). The principal findings are summarized in section Sec. VI.
Finally $B_z S^z_{i,m}$ describes a Zeeman coupling between a local spin-moment and an external longitudinal magnetic field. It is noticeable that the change in sign of $J$ and $D$ do not alter the magnetic ground state as well as excitations above the ground state, whereas $J_z$ and $J_\perp$ are strictly positive in this study.

For $J_\perp \gg |J|$, the ground state of the system is a product of the singlet dimers on each interlayer NN bond. In this limit, the lowest excitations of the system are triplons, which are localized $S = 1$ quasiparticles. An out-of-plane magnetic field lowers the energy of the $S_z = +1$ triplons and at a critical magnetic field, it crosses the energy of the singlet state, populating the ground state with a finite density of triplons. The other triplon branches ($S^2 = 0$ and $S^2 = -1$) are separated by a large energy gap. At low temperatures, one can restrict the local Hilbert space of the dimers to the singlet and $S^2 = +1$ triplon. By treating the triplons as bosonic quasiparticles, one can formulate a description of the low energy physics of the system in terms of hard core bosons which is known as bond-operator formalism (see also Appendix A). In this formalism, the zero field ground state made of singlets on each interlayer bonds is considered an empty lattice with number of triplons $n_t = 0 \forall i$. At the critical field, a finite density of triplons is generated which increases with increasing field. The inter-dimer exchange interactions induce an effective hopping of the triplons. This delocalization induces a BEC of $S_z = +1$ triplons in the ground state. In the spin language, this corresponds to an canted antiferromagnetic order with a spontaneously broken U(1) symmetry. Considering singlets as a vacuum state in the system and triplon ($S_z = +1$) as a hard-core bosonic quasi-particle excitations in vacuum of singlets, we can use the bond operator formalism to express the bi-layer spin Hamiltonian Eq.1 as an effective triplon Hamiltonian on a single-layer honeycomb lattice [15–17] (see Appendix A),

$$\hat{H} = \frac{J}{2} \sum_{\langle i,j \rangle} \left[ \hat{S}^x_i \hat{S}^x_j + \hat{S}^y_i \hat{S}^y_j \right] + \frac{D}{2} \sum_{\langle i,j \rangle} \nu_{ij} \left[ \hat{S}^x_i \hat{S}^x_j - \text{H.c.} \right] + \frac{J_\perp}{4} \sum_i \hat{n}_i \hat{n}_i + J_z \sum_{\langle i,j \rangle} \hat{n}_i \hat{n}_j \tag{2}$$

where, $\hat{t}_i$ is the triplon annihilation operator, $\hat{n}_i$ is the triplon number operator and index-$i$ represents site index of effective single-layer honeycomb lattice (which is equivalent to interlayer bond-index of bi-layer honeycomb lattice). The first two terms represent hopping of triplons between NN and NNN neighbor dimers respectively, the third term is an on-site potential (effectively a chemical potential) and the last term describes the effects of NN interaction between triplons. It is noticeable that the NNN hopping has a complex weight which renders the Hamiltonian unsuitable for quantum Monte Carlo simulations. When $J_z \approx 0$, Eq.2 reduces to a tight binding model of non-interacting triplons. The Bloch Hamiltonian in the momentum basis, in terms of the momentum
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space triplon operators, is determined via Fourier transformation as,

$$\mathcal{H} = \sum_k \Psi_k^\dagger [g(k)\sigma_0 + h \cdot \sigma] \Psi_k, \quad (3)$$

where, \(\Psi_k = (\hat{a}_k, \hat{b}_k)^T\). \(\hat{a}_k (\hat{b}_k)\) denotes the \(k\)-space triplon annihilation operator on sublattice-a (b) as shown in Fig. 1(a)-(b) and \(\sigma\) is the pseudo-vector of Pauli matrices and \(\sigma_0\) is the two-dimensional identity matrix. The coefficients of the \(\sigma\)-matrices in the Bloch Hamiltonian are \(g(k) = (J_\perp/4 - B_z, h_x(k) = (J/2)\sum_i \cos(k \cdot \alpha_i), h_y(k) = (J/2)\sum_i \sin(k \cdot \alpha_i), h_z(k) = D\sum_i \sin(k \cdot \beta_i)\), where \(\alpha_i\) and \(\beta_i\) are the NN and NNN vectors for each layer respectively. \(\beta_i\)'s are chosen such that they form a counter-clockwise triangular loop for sites in sublattice-a in a hexagonal plaquette and clockwise triangular loop for sites in sublattice-b. The energy eigenvalues are given by,

$$E^\pm(k) = g(k) \pm |h(k)|. \quad (4)$$

The band dispersion is shown in Fig. 1(c) and Fig. 1(d) for two values of DMI. For \(D = 0\), the energy spectrum is identical to that of graphene, with a linear band crossing for two values of DMI. For \(D > 0\), the energy spectrum is vanishingly small in our model.

In absence of interaction \(J_z\), Eq. 2 transforms into well-known bosonic topological Haldane model. At finite temperatures due to non-zero magnetic excitations, this model is known to exhibit a finite thermal Hall effect due to presence of non-zero Berry-curve of the bands. On this basis it is expected that Bose-Einstein condensate at \(\Gamma\)-point or \(K (K')\)-point would provide a non-zero Hall conductance even at zero temperature [18]. However, we find that the Berry-curve at the condensation momenta – \(\Gamma\)-point when \(|D| < |J|/\sqrt{3}\) and \(K (K')\)-point when \(|D| > |J|/\sqrt{3}\) – is zero. Since the density of magnons is concentrated around the condensation momentum, and is vanishingly small away from it, the thermal Hall response of the Bose-Einstein condensate is vanishingly small in our model.

An analogous bosonic model can also be observed in terms of Matsubara-Matsuda bosons in magnetically ordered honeycomb ferromagnets (see Fig. 1(b)) or antiferromagnets. Thus the non-trivial phases like twisted-superfluid or twisted-supersolid are also expected to emerge in magnetically ordered systems. However, in practice, the presence of anisotropies in magnetically ordered systems break the \(U(1)\)-symmetries destroying conservation of numbers of particles which in-turn preclude a long time superfluidity response in this systems [15, 21]. Moreover due to presence of strong exchange interactions (\(J, J_z\) in Eq. 2) in general, the phase transitions are also difficult to study varying the external parameters in these systems [21]. That is why the dimerized magnets are well suited to study superfluid phases and superfluid-Mott transitions. Hence we focus on bilayer dimerized honeycomb paramagnetic system to study the twisted superfluid and twisted supersolid phases.
where, \( H \) is the Hamiltonian as in equation Eq.(2) within the cluster and \( H_{BC} \) is the Hamiltonian which takes into account the interactions among the boundary sites of the cluster and the mean-field region. The form of the boundary Hamiltonian is given by,

\[
H_{BC} = \frac{J}{2}\sum_{(i,j)}'\left[\hat{t}_i \langle \hat{t}_j \rangle + \text{H.c.}\right] + \frac{iD}{2}\sum_{(i,j)}'\left[\hat{\delta}_i \langle \hat{\delta}_j \rangle - \text{H.c.}\right] + \frac{J_z}{2}\sum_{(i,j)}'\hat{n}_i \langle \hat{n}_j \rangle ,
\]

where the primed summations are over the boundary site- \( j \) connected to the mean-field site- \( i \). \( \langle \hat{t}_j \rangle \) and \( \langle \hat{n}_j \rangle \) are two mean-field parameters denoting the superfluid order parameter and occupation number of triplons at site- \( j \) respectively. We choose six inequivalent sites in each cluster (denoted by different patterns in figure Fig. 2) to give a total of 12 mean-field parameters. The ground state in the different parameter regimes are obtained by evaluating these mean field parameters self-consistently in the following manner,

(i) Choose an initial set of mean field parameters \( \{ \langle \hat{t}_j \rangle, \langle \hat{n}_j \rangle \} \), \( j = 1, \ldots, 6 \) and then exactly diagonalize the effective Hamiltonian of the cluster \( H_C^{\text{eff}} \).

(ii) Calculate new mean field parameters \( \langle \hat{t}_j' \rangle \) and \( \langle \hat{n}_j' \rangle \) from the sites within blue-dashed rectangle in the figure Fig. 2 which reside within the cluster. Periodic boundary condition is chosen along horizontal direction to eliminate any boundary effect on the sites within the blue-dashed rectangle, so that the mean field parameters obtained from those sites are free from boundary effects.

(iii) The initial and final set of mean field parameters are compared using the tolerance

\[
\epsilon = \sum_j |\langle \hat{t}_j' \rangle - \langle \hat{t}_j \rangle| + \sum_j |\langle \hat{n}_j' \rangle - \langle \hat{n}_j \rangle| .
\]

If the tolerance \( \epsilon \) is less than a certain cutoff then the obtained mean-field parameters correspond to the ground state of the system. Otherwise the step (i) is repeated with new values of mean-field parameters \( \langle \hat{t}_j \rangle = \langle \hat{t}_j' \rangle \) and \( \langle \hat{n}_j \rangle = \langle \hat{n}_j' \rangle \).

We set the cutoff as \( 10^{-10} \) and start the simulation with different initial mean-field parameter sets for a fixed set of parameters \( J, D, J_1, B_z, \) and \( J_z \). In general, the simulations with different initial mean field parameter sets give different ground-states at the boundary of two phases and we selected the phase with minimum energy as the ground state.

After obtaining the ground state via self-consistent determination of the mean field parameters, four order-parameters are calculated to identify the nature of the ground state phase of the system. The magnitude of superfluid order parameter is given by,

\[
|b| = \max \left| \langle \hat{t}_j \rangle \right| .
\]
where \( \text{max} \) denotes maximum value of the parameter obtained out of six-sites within the dashed blue-border in Fig. 2. Additionally, the average number of particles per site \( \langle n \rangle_{av} \) and difference in number of particles between NN sites \( \Delta n \) are also enumerated. The superfluid order-parameter is a complex quantity and for twisted superfluid phases in our study the phase difference of superfluid order parameter \( \theta \) among NNN sites is obtained to be \( \theta = 120^\circ \) and otherwise \( \theta = 0^\circ \).

IV. NUMERICAL RESULTS

![Fig. 3: (Color online) The order parameters are plotted for parameter values (a) \( D = 0.2J, J_z = J \), (b) \( D = 0.9J, J_z = J \), (c) \( D = 0.2J, J_z = 4J \), (d) \( D = 0.9J, J_z = 4.5J \).](image)

Using CGMFT, we determine the order parameters \( |b| \), \( \langle n \rangle_{av} \), \( \Delta n \) and \( \theta \) as a function of magnetic field \( B_z \) for different sets of the parameters \( (D, J_z) \). The evolution of the order parameters and the resulting field driven phases are shown in Figure fig. 3 for four illustrative points of the \( (D, J_z) \) parameter space. In fig. 3, the DMI increases from the left-column of figures to the right-column of figures, whereas the interaction \( J_z \) increases from upper-row of figures to the lower-row of figures.

For weak DMI \( (D = 0.2J) \), the field driven phase diagram resembles that of the canonical extended Bose Hubbard model for hard core bosons [40] (see Fig. 3(a), (c)). The zero field \( (B_z = 0) \) ground state corresponds to a singlet phase, or equivalently an empty lattice in the bosonic language. All the order parameters vanish in this limit. This remains true at small values of the applied field reflecting a finite gap to lowest excitations due to the singlet-triplet gap of the local dimers. When the applied field exceeds a critical value, the gap is closed and the ground state acquires a finite density of triplons. These field induced triplons form a superfluid (SF) driven by the NN triplon hopping and is characterised by a finite SF order parameter, \( |b| \). The mismatch between the occupancy of the two sublattices \( \langle \Delta n \rangle \) remains zero, reflecting the uniform nature of the SF phase. A vanishing twist angle \( (\theta = 0) \) completes the characterization of the phase as a normal superfluid. In the weak interaction limit \( (J_z < 2J) \) with increasing magnetic field, the density of triplons increases monotonically till full saturation is reached at an upper critical field when each dimer is occupied by a triplon. At saturation, all the order parameters (except average density, \( \langle n \rangle_{av} \), of triplons) vanish denoting a lattice fully occupied by triplons. In this weak interaction limit, the physics is similar to the non-interacting limit as described in section II and so the qualitative feature can be well described using band structure in figure Fig. 1(c).

For strong interactions \( (J_z > 2J) \), an intervening charge density wave (CDW) phase, driven by the strong NN-interaction between triplons, appears in addition to the phases discussed above (see Fig. 3(c)). With increasing magnetic field, when the density of triplons reaches \( \langle n \rangle_{av} = 1/2 \), the triplons form a staggered CDW pattern where one of the sublattices is fully occupied, while the other remains empty. The potential energy cost due to nearest neighbor interaction is minimized as there are no nearest neighbor pairs. This is accompanied by a complete quenching of superfluidity, since any hopping of triplons will necessarily involve configurations with energetically costly multiple nearest neighbor pairs. The CDW phase has a finite gap to the addition of any more triplons and the density remains constant at \( \langle n \rangle_{av} = 1/2 \) over a finite range of applied field. This phase is characterized by a vanishing superfluid order, and a non-zero density mismatch between the two sublattices \( \langle \Delta n \rangle \), reflecting the staggered order. When the increasing field strength reaches a critical value where the Zeeman energy gain due to increasing magnetization (equivalently, adding more triplons) exceeds the potential energy cost of nearest neighbor repulsion, the density of triplons starts to increase again, resulting in another normal SF phase. Finally, as the field is increased above a saturation value, \( B_{sat} \), the ground state enters the fully polarized phase.

The above argument for the appearance of interaction driven CDW phase at half-filling does not apply for weak to moderate interaction strengths \( (J_z < 2J) \), as the kinetic energy gain due to the delocalization of triplons exceeds the potential energy cost of NN-interactions.

The sequence of field-driven phase changes markedly for strong DMI. As shown earlier in Sec. II, in the non-
interacting limit, the triplon band minimum shifts from the Γ point to the K and K’ (Fig. 1(d)) and the BEC of triplons occur at finite momentum. A local minimum persists at the center of the Brillouin zone, and the energy gap between the triplon-sector and the singlet dimer sector, $E_K$ in Eq. 5 decreases with increasing DMI. This behavior persists in the presence of weak to moderate interaction ($J_z < 2J$) and is reflected in Fig. 3(b). For the present choice of parameters, the energy of the lowest triplon excitation is vanishingly small. The triplon density acquires a finite value for an infinitesimally small $B_z$, and increases monotonically with the strength of the applied field. In this regime, the triplons form a superfluid ($|b| > 0$). More interestingly, the complex NNN hopping process imparts a complex phase to the superfluid order parameter, as seen by a finite expectation value of the twist angle ($\theta \neq 0$). In other words, the ground state in this parameter range is a twisted superfluid (TSF). The triplon density increases monotonically, with the ground state remaining a TSF, till the fully polarized phase is reached at a saturation field, $B_{sat}$.

Finally in the strong DMI and strong interaction limit (see Fig. 3(d)), the twisted superfluid is replaced by a twisted supersolid phase, in addition to the appearance of an interaction-driven CDW phase at $\langle n \rangle_{av} = 1/2$ over a finite range of applied field. In the twisted supersolid phase (TSS), the ground state is characterized by a finite $\Delta n$ (density mismatch between the two sublattices), in addition to a complex superfluid order parameter ($|b| \neq 0, \theta \neq 0$). The finite density difference between two sublattices provide the diagonal order concurrently with the finite (twisted) superfluid ordering. It is surprising that the ground state exhibits TSS order even at low triplon densities. This is understood by recalling that the primary delocalization process in this parameter regime involves the DMI-induced intrasublattice complex next-nearest-neighbor hopping. The strong NN-repulsion between the triplons further suppresses inter-sublattice hopping processes, resulting in a preferential occupation of one of the two sublattices at small densities.

The phase diagram in $B_z - D$ parameter space is shown in the figure Fig. 4 for two different values of interaction $J_z = J$ and $J_z = 4.5J$. A comparison of the phase diagrams at moderate ($J_z < 2J$) and strong ($J_z > 2J$) interactions reveal,

(i) Appearance of CDW phase at half-filling in the strong interaction limit. The CDW phase appears at half filling dividing the SF-region which appear at weak to moderate $J_z$ into two SF-regions.

(ii) In the strong interaction limit the TSF phase is replaced by a TSS phase.

V. MATERIAL REALIZATION

There are two main ingredients to realize TSF or TSS phases; firstly, a bilayer honeycomb valence bond state is required; secondly, a DMI greater than a critical value is required. Although, to the best of our knowledge, there are no materials described in the literature which satisfies our model, a rough idea is sketched here to obtain TSS or TSF phases; firstly, a bilayer honeycomb valence bond state is required. Although, to the best of our knowledge, there are no materials described in the literature which satisfies our model, a rough idea is sketched here to obtain TSS or TSF phases; firstly, a bilayer honeycomb valence bond state is required. Although, to the best of our knowledge, there are no materials described in the literature which satisfies our model, a rough idea is sketched here to obtain TSS or TSF phases. Secondly, a DMI greater than a critical value is required. 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The material Bi$_3$Mn$_4$O$_{12}$(NO$_3$)$_3$ consists of Mn$^{4+}$ ions carrying spin $S = \frac{3}{2}$ arranged in a bilayer honeycomb lattice with A-A stacking. No magnetic ordering is observed down to the lowest temperatures. Theoretical studies indicate that the disordered magnetic state may be an interlayer dimer phase which is adiabatically connected to the direct products of singlets [41–45], although a spin liquid phase cannot be completely ruled out [46–57]. If the ground state phase is a spin liquid, an interlayer dimer phase can be induced by applying a pressure along the perpendicular direction [44, 51].

On the other hand, experimentally well studied van der Waals material CrI$_3$ is a honeycomb ferromagnet where Cr$^{2+}$ ions carry spin-$\frac{1}{2}$ moment forming a honeycomb lattice. This material has been well studied by tuning the number of layers [58] as well as varying the
pressure [59]. It is shown that the interlayer antiferromagnetic Heisenberg exchange interaction changes linearly with the distance between two layers by application of pressure on the material and can be achieved by a interlayer coupling twice as compared with the initial interlayer coupling [59]. Although a valence bond state is not detected in CrI$_3$ (and there are indications of structural phase transition under pressure) [59], the pressure induced high interlayer coupling in CrI$_3$ motivates for searching of pressure induced valence bond states in a family of Van der Walls honeycomb magnets CrBr$_3$ [60], CrGeTe$_3$ [61], CrSiTe$_3$ [62], FePS$_3$ [63], NiPS$_3$ [64].

Another challenge for achieving the TSF and TSS phases is to obtain a high DMI (presence of frustration among interlayer and intralayer of DMI to realize TSF or TSS states is reduced due to light may not be required for a real material if the cutoff frequency of the light nearby $\nu$ is based on single-band extended Hubbard model; for a magnetic material which cannot be described in Ref. [67] is based on single-band extended Hubbard. The scalar spin chirality as in reference Ref. [67] given by, $\chi_3 = \sum_{\langle j,k \rangle} \nu_{jk} \langle \tilde{n}_i \rangle \left[ \tilde{t}_{j,k}^\dagger - H.c. \right]$, (10)

where the $i$-th site is the NNN neighbor of both $j$-th and $k$-th sites, $\chi$ is synthetic scalar spin chirality. Comparing the equations Eq. 2 and Eq. 10, the effective DMI for bond $\langle j,k \rangle$ due to circularly polarized light is $D = 3\chi/4$. The scalar spin chirality $\chi$ is shown to have a resonance for the frequency of light near $\omega_n = 1/n^2$, where $U$ is onsite interaction representing electron-electron repulsion and $n$ is a positive integer. Thus the effective DMI can be tuned as high as possible tuning the frequency of the light nearby $\omega_n$. However the results in Ref. [67] is based on single-band extended Hubbard model; for a magnetic material which cannot be described by a one band Hubbard model, one needs a more careful theoretical treatment. On the other hand, application of light may not be required for a real material if the cutoff of DMI to realize TSF or TSS states is reduced due to presence of frustration among interlayer and intralayer interactions (see Appendix. C).

VI. CONCLUSION

To summarize, we have shown that a magnetic analog of the novel twisted superfluid (TSF) state reported in recent experiments with ultracold atoms in an optical lattice [1] can be realized in a bilayer quantum antiferromagnet with realistic interactions. We show that

TSF phase is induced by DMI greater than a critical value. For Ising-like anisotropy of the intra-plane Heisenberg interactions, the TSF phase is replaced by a twisted supersolid (TSS) phase. While the strength of DMI required for the stabilization of TSF and TSS phases ($D/J > 0.5$) is not observed natively in most quantum magnets, recent experiments have shown that a strong DMI can be induced in thin films of insulating magnets, by forming heterostructures with heavy metals (with strong spin-orbit coupling) [69–72]. Our results show that circularly polarized light can also induce large DMI by varying the frequency of light [67]. Finally, the presence of frustration among interlayer and intralayer Heisenberg exchange interactions in a material can lower the value of the critical DMI required to realize TSF and TSS phases (Appendix. C), thus facilitating their experimental observation. We propose that the material Bi$_3$Mn$_4$O$_{12}$ (NO$_3$) [41–45] and family of honeycomb magnets CrBr$_3$ [60], CrGeTe$_3$ [61], CrSiTe$_3$ [62], FePS$_3$ [63], NiPS$_3$ [64] are promising candidate materials to realize TSF and TSS phases.

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Appendix A: Bond-operator formalism

The spin-operators in terms of bond-operators are given by [17],

$$\hat{S}_{j,l}^+ = \frac{\hat{t}_{1,j,l}^\dagger \hat{t}_{0,j,l} + \hat{t}_{0,j,l}^\dagger \hat{t}_{1,j,l}}{\sqrt{2}} \pm \frac{\hat{s}_j^\dagger \hat{t}_{1,j} - \hat{t}_{1,j}^\dagger \hat{s}_j}{\sqrt{2}}$$

$$\hat{S}_{j,l}^- = \frac{\hat{t}_{1,j,l}^\dagger \hat{t}_{0,j,l} + \hat{t}_{0,j,l}^\dagger \hat{t}_{1,j,l}}{\sqrt{2}} \pm \frac{\hat{s}_j^\dagger \hat{t}_{1,j} - \hat{t}_{1,j}^\dagger \hat{s}_j}{\sqrt{2}}$$

$$\hat{S}_{j,l}^{z} = \frac{\hat{t}_{1,j,l}^\dagger \hat{t}_{1,j} - \hat{t}_{1,j}^\dagger \hat{t}_{1,j}^\dagger}{2} \pm \frac{\hat{s}_j^\dagger \hat{t}_{0,j} + \hat{t}_{0,j}^\dagger \hat{s}_j}{2},$$

where the upper-sign is for layer $l=A$ and the lower sign is for the layer $l=B$. $\hat{t}_{1,j,l}$ and $\hat{t}_{0,j}$ create states $|\uparrow\uparrow\rangle$ and $(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle)/\sqrt{2}$ on $j$-th bond respectively. After the bond operator transformation, the bilayer honeycomb spin system transforms into an effective single layer honeycomb lattice system with triplon and singlet operators on each site. At low temperature, in the limit $|J_{\perp}| \gg |J|$, it can be assumed that the ground state is...
product of singlets on the interlayer NN-bonds. Assuming singlets form the empty or vacuum state and taking advantage of the hardcore nature of the bosons, we can simply transform the following quadratic operators into a single triplon operators in equation Eq. A1,

\[ s^z_j \hat{t}_{\alpha j} \rightarrow \hat{t}_{\alpha j}, \quad s^z_j \hat{\tilde{t}}_j \rightarrow \hat{\tilde{t}}_j. \]

Moreover application of magnetic field in the z-direction lowers the energy of triplon correspond to the operator \(\hat{t}_j\) and so neglecting all other triplon operators in equation Eq. A1, we get the following bond operator transformation [15],

\[ \hat{S}^+_{j,l} = \frac{1}{\sqrt{2}} \hat{t}_j, \quad \hat{S}^-_{j,l} = \frac{1}{\sqrt{2}} \hat{\tilde{t}}_j, \quad \hat{S}^z_{j,l} = \frac{1}{2} \hat{t}_j, \]

thus coexistence of particles at \(\mathbf{K}\) and \(\mathbf{K}'\) points does not seem to increase the energy of the system. However, quantum fluctuations around the points \(\mathbf{K}\) and \(\mathbf{K}'\) increases the energy due to presence of triplons at both \(\mathbf{K}\) and \(\mathbf{K}'\) points as shown in the reference Ref. [18].

\[ \mathcal{H}_V = \frac{3J}{2} \left[ \hat{\mathbf{t}}_{\mathbf{K}} \cdot \hat{\mathbf{t}}_{\mathbf{K}'} + \hat{\tilde{t}}_{\mathbf{K}} \cdot \hat{\tilde{t}}_{\mathbf{K}'} + \hat{\mathbf{t}}_{\mathbf{K}} \cdot \hat{\tilde{t}}_{\mathbf{K}'} + \hat{\tilde{t}}_{\mathbf{K}} \cdot \hat{\mathbf{t}}_{\mathbf{K}'} \right] \\
+ \frac{J_z}{2} \sum_{\alpha} \left[ \epsilon^{(K-K')} \alpha \hat{\mathbf{t}}_{\mathbf{K}} \cdot \hat{\mathbf{t}}_{\mathbf{K}'} + \epsilon^{(K-K')} \alpha \hat{\tilde{t}}_{\mathbf{K}} \cdot \hat{\mathbf{t}}_{\mathbf{K}'} + \epsilon^{(K-K')} \alpha \hat{\mathbf{t}}_{\mathbf{K}} \cdot \hat{\tilde{t}}_{\mathbf{K}'} + \epsilon^{(K-K')} \alpha \hat{\tilde{t}}_{\mathbf{K}} \cdot \hat{\tilde{t}}_{\mathbf{K}'} \right] \]

\[ = \frac{3J_z}{2} \left( \hat{n}_{\mathbf{K}} + \hat{n}_{\mathbf{K}'} \right) + \frac{3J_z}{2} \left( \hat{n}_{\mathbf{K}} + \hat{n}_{\mathbf{K}'} \right)^2, \quad (B2) \]

\[ \mathcal{H} = J_1 \sum_{i,m \in A} \mathbf{S}_{i,m} \cdot \mathbf{S}_{i,n} - D_z \sum_{i,m} S^z_{i,m} + \sum_{\langle i,j \rangle, m} [J(S^z_{i,m} S^z_{j,m} + S^y_{i,m} S^y_{j,m}) + J_z (S^z_{i,m} S^z_{j,m})] \]

where the subscript-1 is omitted from the operator \(\hat{t}_{1,j}\).

**Appendix B: Interaction term in reciprocal space**

The last term in equation Eq. 2 represents the NN repulsive interaction and in reciprocal space it is given by,

\[ \mathcal{H}_V = \frac{J}{2} \sum_{\alpha} \sum_{\mathbf{k}, \mathbf{k}', \nu} \epsilon^{(K-K')} \alpha \hat{\mathbf{t}}_{\mathbf{K}'} \cdot \hat{\mathbf{t}}_{\mathbf{K}}. \quad (B1) \]

**Appendix C: Introducing more terms in the spin Hamiltonian**

In this appendix, we have taken into account additional interlayer bonds (see Fig. 5(a), (b)). We introduce intralayer NNN Heisenberg-exchange interaction \(J_1\) as well as interlayer NN Heisenberg exchange interaction \(J_{2,\bot}\) in the spin Hamiltonian in equation Eq. 1,

\[ \mathcal{H} = J_1 \sum_{i,m \in A} \mathbf{S}_{i,m} \cdot \mathbf{S}_{i,n} - D_z \sum_{i,m} S^z_{i,m} + \sum_{\langle i,j \rangle, m} [J(S^z_{i,m} S^z_{j,m} + S^y_{i,m} S^y_{j,m}) + J_z (S^z_{i,m} S^z_{j,m})] \]

Moreover we have added symmetry allowed interlayer NNN DMI \(D_{\bot}\) and Heisenberg interaction \(J_{3,\bot}\). The DMI on dimer-bond and NN interlayer bonds are zero due to presence of inversion center at the middle of the bonds. The
corresponding real space triplon Hamiltonian is given as,

\[ H = \frac{J - J_{2\perp}}{2} \sum_{\langle ij \rangle} \left[ \tilde{t}_i \tilde{t}_j + \text{H.c.} \right] + \frac{J_1 - J_{3\perp}}{4} \sum_{\langle \langle ij \rangle \rangle} \left[ \tilde{t}_i \tilde{t}_j + \text{H.c.} \right] + \frac{i(D + D_{\perp})}{2} \sum_{\langle \langle ij \rangle \rangle} \nu_{ij} \left[ \tilde{t}_i \tilde{t}_j - \text{H.c.} \right] + \left( \frac{J_{\perp}}{4} - B_z \right) \sum_i \tilde{t}_i^\dagger \tilde{t}_i + \frac{J_{2\perp} + J_1}{2} \sum_{\langle ij \rangle} \hat{n}_i \hat{n}_j + \frac{J_1 + J_{3\perp}}{4} \sum_{\langle \langle ij \rangle \rangle} \hat{n}_i \hat{n}_j \]  

(C2)

It is noticeable that the Heisenberg interaction \( J_{2\perp} \) renormalizes the interactions \( J \) and \( J_1 \), whereas \( J_1 \) and \( J_{3\perp} \) add additional NNN hopping term and NNN interaction term. Moreover \( D_{\perp} \) renormalizes \( D \) by simply adding up and so \( D_{\perp} \) is absorbed into \( D \) in the rest of this section. In this appendix, we investigate the triplon Hamiltonian, neglecting the interaction terms. The non-interacting Hamiltonian in \( k \)-space is similar to equation Eq. 3 and given by,

\[ H_0 = \sum_k [g'(k) \sigma_0 + h'(\sigma)] \Psi_k, \]

(C3)

where,

\[ g'(k) = (J_{\perp}/4) - B_z + ((J_1 - J_{3\perp})/2) \sum_i \cos(k \cdot \alpha_i), \]

\[ h'_x(k) = ((J - J_{2\perp})/2) \sum_i \cos(k \cdot \beta_i), \]

\[ h'_y(k) = ((J - J_{2\perp})/2) \sum_i \sin(k \cdot \alpha_i), \]

\[ h'_z(k) = D \sum_i \sin(k \cdot \beta_i), \]

where \( \alpha_i \) and \( \beta_i \) are the NN and NNN vectors for each layer respectively. The band structure for two different \( D \)-values are plotted in the figure Fig. 5(c) and (d). The minima of the bands are at the \( \Gamma \)-point and \( K \) (or \( K' \))-point for \( D = 0.1J \) and \( D = 0.8J \) respectively. The results are same when \( J_{2\perp} = 0 \) and \( J_1 = 0 \) as in the figures Fig. 1(c) and (d). The condition for the band minima at \( K \) or \( K' \)-point is given as,

\[ |D| > \frac{2|J - J_{2\perp}| - 3(J_1 - J_{3\perp})}{2\sqrt{3}}. \]

(C4)

We note that the critical DMI required to realise TSF or TSS phases depends on various Heisenberg exchange interactions. The conditions \( J \approx J_{2\perp} \) and \( J_1 \approx J_{3\perp} \) lowers the critical value of the DMI to realize TSF or TSS phases. Thus we can conclude that frustration among interlayer and intralayer Heisenberg interactions can lower the critical DMI required to realize TSF and TSS phases.

![Diagram](image-url)

FIG. 5: (Color online) (a) Intradimer and interlayer NN bonds are indicated by red solid and red dotted lines, respectively. (b) The intralayer and interlayer NNN bonds are indicated by green solid and green dotted lines, respectively. (c) Triplon band structure at \( D = 0.1J \), (b) Triplon band structure at \( D = 0.8J \). The other parameters for the band structures are \( J_{\perp} = 10J \), \( J_1 = 0.8J \), \( J_{2\perp} = 3J \), \( B_z = 0.0 \), \( J_z = 0.0 \), \( J_{3\perp} = 0.0 \).

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