

Supplementary Materials: Evidence for field induced quantum spin liquid behavior in a spin-1/2 honeycomb magnet

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S1. HONEYCOMB LATTICE

The geometry of the Honeycomb layer in NCTO is shown in Fig.S1, where Fig.S1(a) illustrates the bases $\mathbf{a}$, $\mathbf{b}$ of translation as well as the spin axes x, y, z , and Fig.S1(b) shows the coordinates of various directions in the $\mathbf{a}^*, \mathbf{b}^*, \mathbf{c}^* = \frac{2\pi}{|c|} \mathbf{c}$ bases (black) and in the x, y, z bases (red) respectively.

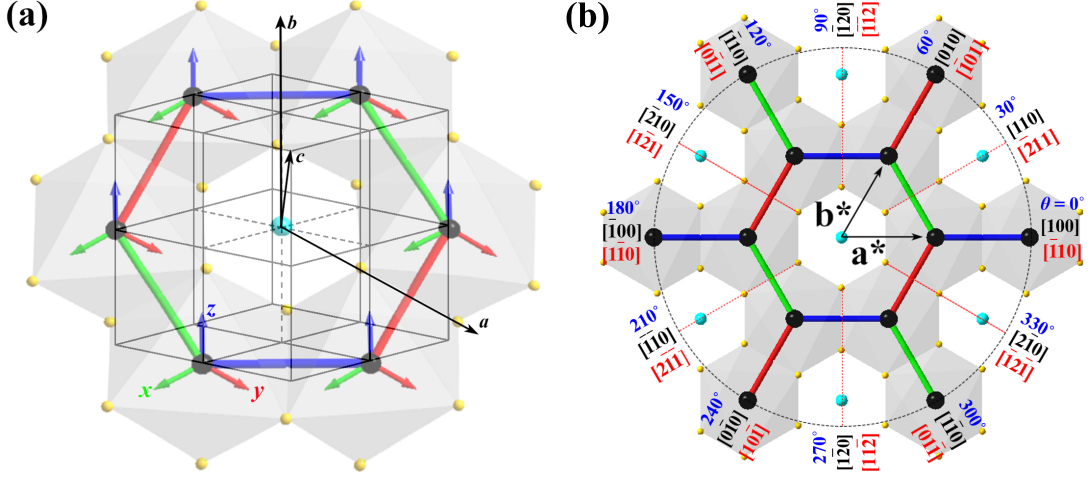


FIG. S1: **Schematic of the crystal structure of Na₂Co₂TeO₆.** (a) Definitions of the crystallographic axes (a , b , c) and the Ising spin axes (x , y , z). (b) Honeycomb network showing the Co-Co bonds (red/blue/green) and CoO₆ octahedras (the black, cyan and golden spheres represent Co, Te and O atoms, respectively). The black arrows are the two principal reciprocal vectors $\mathbf{a}^*$ and $\mathbf{b}^*$ in first Brillouin zone. Magnetic field $\mathbf{B}$ is applied within the honeycomb plane. θ is the angle between $\mathbf{B}$ and $\mathbf{a}^*$ and with respect to the real-space orientation of the Co-Co bonds. The black and red fonts represent the coordinate vector in reciprocal space and spin-axes coordinate, respectively. Directions that are equivalent to $[1,0,0]$ and $[1,1,0]$ correspond to angles $\theta = n \times 60^\circ$ and $\theta = n \times 60^\circ + 30^\circ$, respectively (where n is an integer) in reciprocal space coordinate.

S2. MAGNETIZATION AND SPECIFIC HEAT

The single-crystal DC magnetic susceptibility $\chi(T)$, Fig. S2(a), measured by the zero-field cooling at 0.01 T and the single-crystal specific heat $C_P(T)/T$ at 0 T, Fig. S2(b), show two sharp anomalies at $T_N \approx 26$ K with zigzag antiferromagnetic order and $T_F \approx 15$ K with a spin reorientation transition in the ab plane. A weak anomaly of the $\chi(T)$ and $C_P(T)/T$ curves can be also seen around $T^* \approx 7$ K, which may be related to the modulation of magnetic domain. Their values mirror well the previous reports[1–5]. Due to the magnetic-exchange anisotropy in NCTO, the magnetic order state can observe a distinct magnetic anisotropic behavior along the different crystal-axis directions, Fig. S2(a). However, the $\chi(T)$ curves also present a strongly anisotropic behavior above T_N in the ab plane and c -axis, which is mainly attributed to the strongly anisotropic g factor originating from the low-spin state of Co²⁺. Hence, the spin-1/2 model is an effective, low-energy description for the Co²⁺ ions in the temperature range considered in this work, which is consistent with our previous report[5].

In order to better clarify the magnetic phase diagram in the main text, we perform isothermal magnetization $M(B)$ with the applied magnetic field $\mathbf{B} // \mathbf{a}^*$ -axis at selected temperatures, Fig. S2(c) and (d). The critical fields B_{C1} and B_{C2} are comparable with results of the magnetic torque.

Figure S2(e) shows the magnetic specific heat C_{mag}/T curves with $\mathbf{B} // \mathbf{a}^*$ -axis, subtracting phonon contributions measured on an isostructural Na₂Zn₂TeO₆ reference crystal that is shown in the Fig. S2(b). The critical temperatures in Fig. 1(a) in the main text is marked by the anomalies in the C_{mag}/T curves.

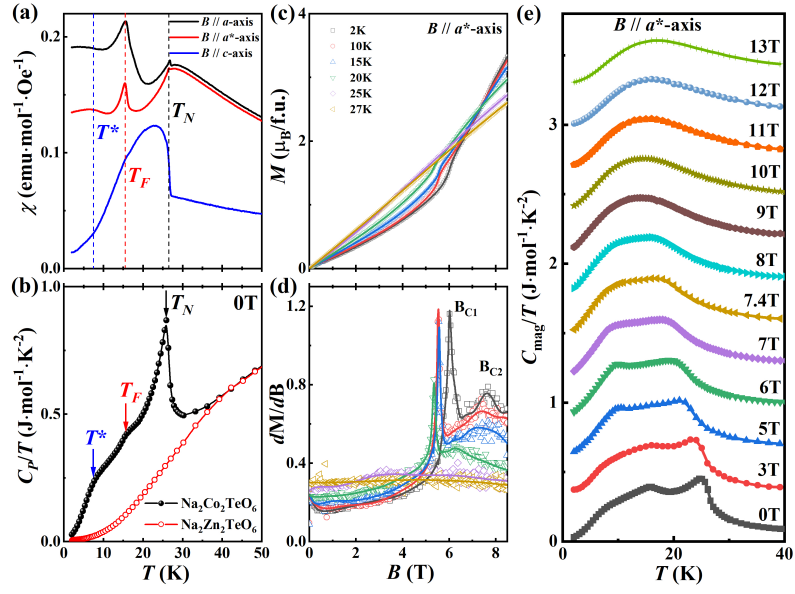


FIG. S2: **Macroscopic physical properties of $\text{Na}_2\text{Co}_2\text{TeO}_6$.** (a) Temperature dependence of susceptibility $\chi(T)$ in NCTO measured at 0.01 T along the different crystal-axis directions. (b) Temperature dependence of specific heat under 0 T for NCTO and $\text{Na}_2\text{Zn}_2\text{TeO}_6$. (c) The isothermal magnetization $M(B)$ with the applied magnetic field $\mathbf{B} \parallel \mathbf{a}^*$ -axis at selected temperatures. (d) The differential isothermal magnetization as functions of fields dM/dB vs. B , originating from the data of (c). The solid lines represent the fitted curves by polynomial in (c) and (d). The critical fields B_{C1} and B_{C2} are determined by the dM/dB curves, which are comparable with the critical fields in the main text. (e) Temperature dependence of magnetic specific heat in NCTO under selected fields.

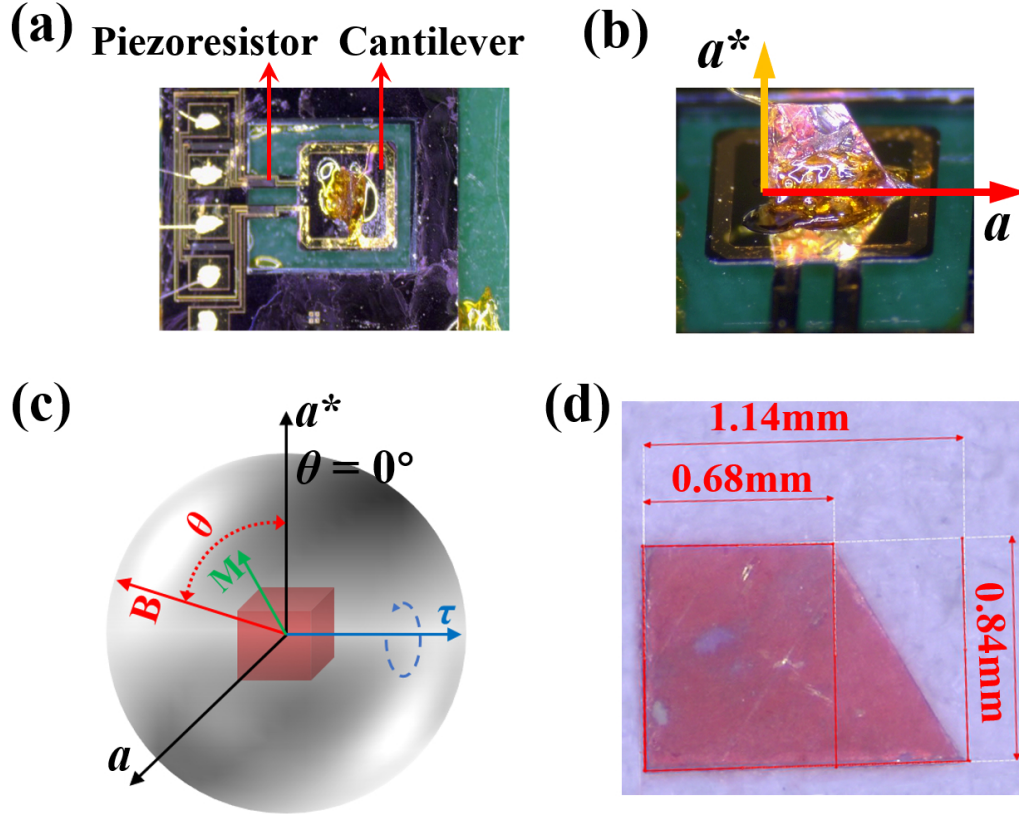


FIG. S3: **Set up of magnetic torque measurements.** (a) and (b) illustrate the experimental configuration for angle-dependent magnetic torque $\tau(\theta)$ measurements. (c) The polar angular relations between the $\mathbf{a}^*$ and $\mathbf{B}$ in the rotated plane. (d) Morphology and size of the selected sample.

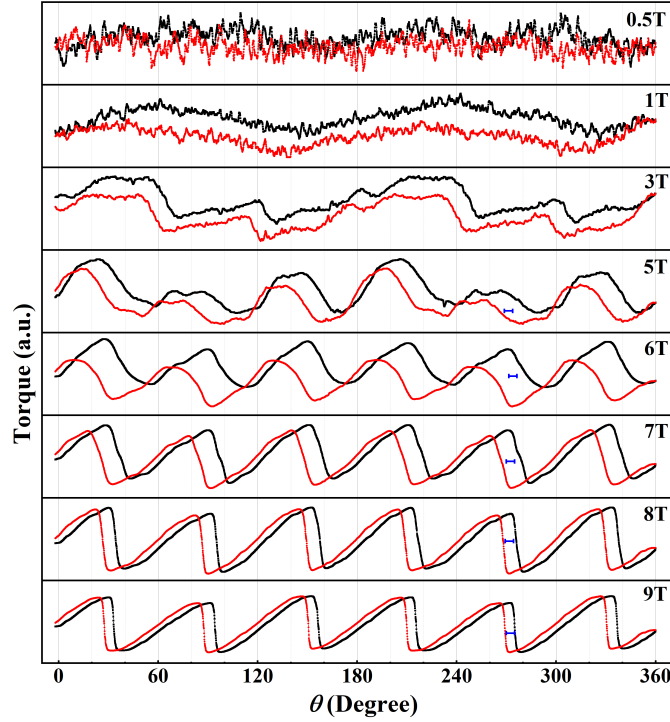


FIG. S4: **Angle-dependent magnetic torque without subtracting background at 2 K with different fields.** The blue horizontal error bars indicate the instrumental backlash (~ 5 degrees) when reversing rotation. The black solid lines represent the increasing angles from 0 to 360 degrees. The red solid lines represent the decreasing angles from 360 to 0 degrees.

S3. MAGNETIC TORQUE

Figure S4 shows the angle-dependent magnetic torque at 2 K. The backlash of the mechanical rotation is about 5 degree, marked by the blue horizontal error bars in Fig. S4. Therefore, we observe that the angle-dependent magnetic torque curves will have hysteresis obtained by counterclockwise and clockwise rotations. Surprisingly, a larger direction deviation of more than 5 degrees is seen between about 5 T and 7 T, which cannot be attributed to the error caused by the instrument. We note that an obvious hysteresis can be seen in the field dependence of magnetic torque and magnetization, Fig. S5, indicating the first-order phase transition around B_{C1} that is just between 5 T and 7 T. Hence, such large angular deviation of Fig. S4 between about 5 T and 7 T should be related to the first-order phase transition around B_{C1} .

In order to draw the magnetic phase diagram in the Fig. 1(a) of the main text, we measure the field dependence of magnetic torque along the angle $\theta = 10.6^\circ$ at different temperatures, Fig. S6. The superimposed color map of Fig. 1(a) is from the differential $\frac{1}{B} \frac{d\tau}{dB}$ of Fig. S5.

S4. INELASTIC NEUTRON SCATTERING

Figures S7(a) and (b) show the rod-like dependence on the out-of-plane momentum component L , demonstrating the inter-layer interaction is relatively weak, NCTO could be regarded as a 2D Co^{2+} -based honeycomb magnet.

S5. VARIATIONAL MONTE CARLO SIMULATIONS

A. For the Ground States

The VMC approach is based on the spinon representation by introducing two species of fermions $C_i^\dagger = (c_{i\uparrow}^\dagger, c_{i\downarrow}^\dagger)$. The spin operators are written in quadratic forms of the fermionic spinons $S_i^m = \frac{1}{2} C_i^\dagger \sigma^m C_i$ under the particle

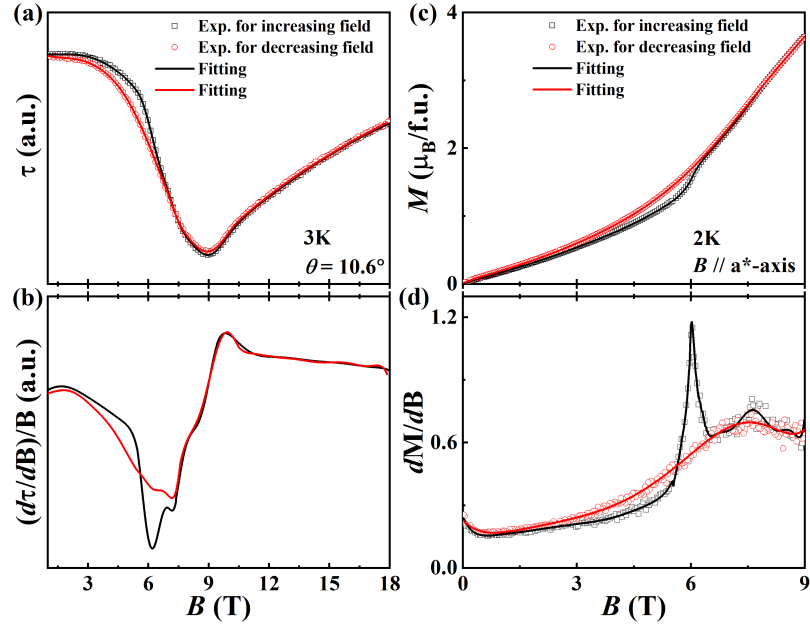


FIG. S5: **Field dependence of magnetic torque and magnetization.** Field dependence of magnetic torque (a) and differential magnetic torque (b) in NCTO measured at 3 K with magnetic field 10.6 degrees away from a^* -axis. Field dependence of magnetization (c) and differential magnetization (d) in NCTO measured at 2 K along the field $B \parallel a^*$ -axis.

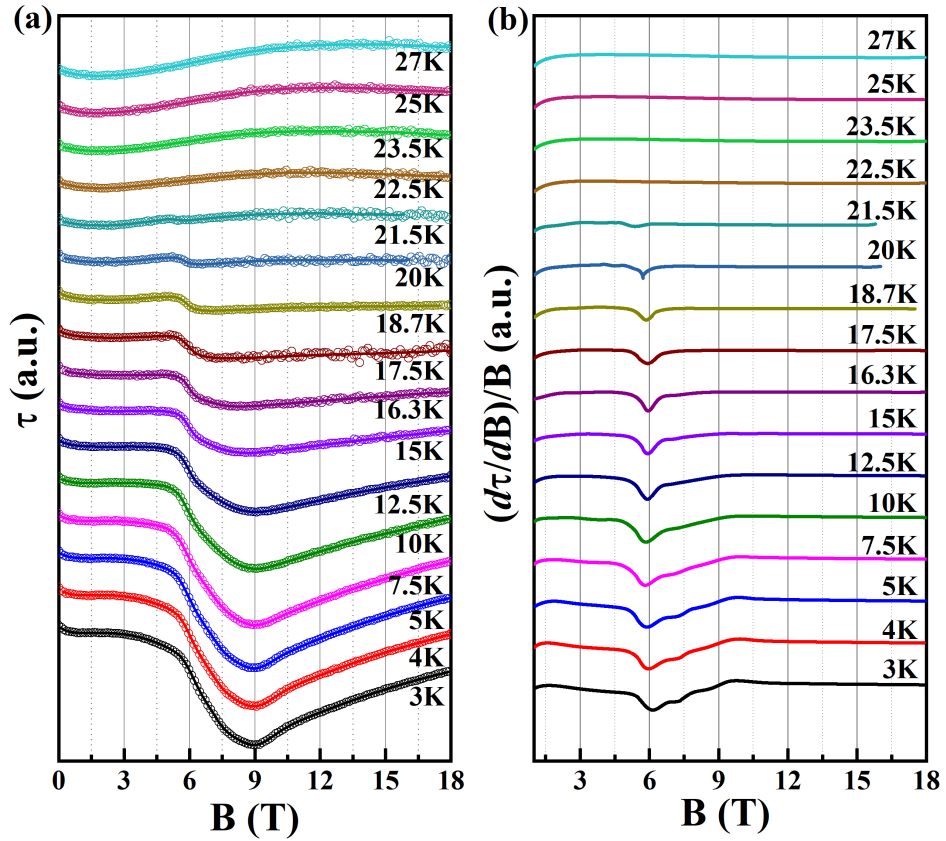


FIG. S6: **Isothermal magnetic torque with field close to a^* -axis.** Field dependence of magnetic torque (a) and differential magnetic torque (b) in NCTO measured at different temperatures along the angle $\theta = 10.6^\circ$.

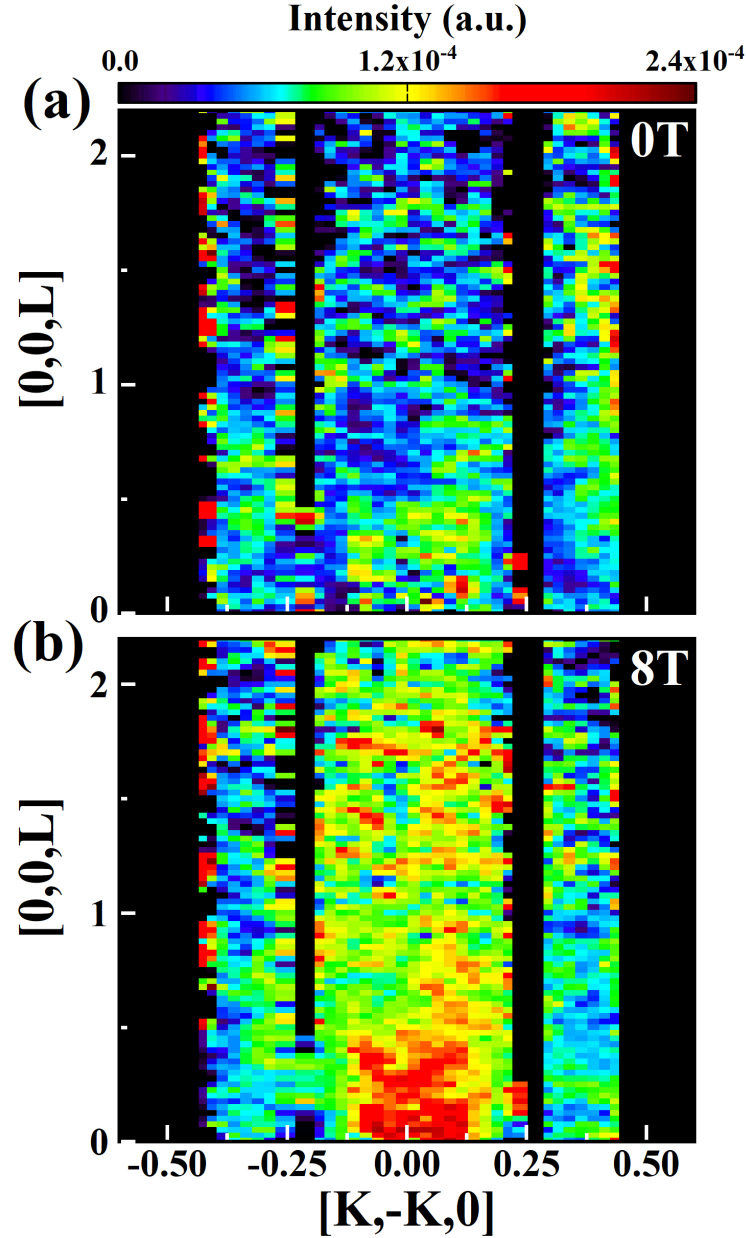


FIG. S7: **Rod-like dependence on the out-of-plane momentum component L .** (a) and (b) Constant-energy scattering at 0 T and 8 T, respectively, integrated over $H = [-0.1, 0.1]$ and $E = [1.5, 2.5]$ meV, projected on the reciprocal honeycomb plane defined by the perpendicular directions $[0, 0, L]$ and $[K, -K, 0]$. The color bar indicates scattering intensity in arbitrary unit in linear scale. The black regions lack detector coverage.

number constraint, $\hat{N}_i = c_{i\uparrow}^\dagger c_{i\uparrow} + c_{i\downarrow}^\dagger c_{i\downarrow} = 1$, where $m \equiv x, y, z$, and σ^m is Pauli matrix. It is convenient to introduce the matrix operator $\psi_i = (C_i, \bar{C}_i)$ with $\bar{C}_i = (c_{i\downarrow}^\dagger, -c_{i\uparrow}^\dagger)^T$ such that the spin operators can also be written as $S_i^m = \text{Tr}(\psi_i^\dagger \frac{\sigma^m}{4} \psi_i)$. Since the spin operators are invariant under a local $SU(2)$ transformation $\psi_i \rightarrow \psi_i W_i$, the fermionic spinon representation has an $SU(2)$ gauge ‘symmetry’[6].

The spin interactions are rewritten in terms of interacting fermionic operators and are further decoupled into a non-interacting mean-field Hamiltonian. In a quantum spin liquid (QSL) phase, all the physical symmetries are preserved in the ground state, and the $SU(2)$ gauge symmetry may reduce to $U(1)$ or Z_2 . For instance, the gauge symmetry of the Kitaev spin liquid is Z_2 . The mean-field Hamiltonian of a QSL phase does not necessarily preserve the physical symmetries, but should be invariant under an extended group called the ‘projective symmetry group’ (PSG)[7]. We adopt the same PSG of the Kitaev spin liquid[8, 9], and the corresponding QSL mean field Hamiltonian

of the K - J_1 - Γ - Γ' - J_3 model reads

$$\begin{aligned}
H_{\text{mf}}^{\text{QSL}} = & \sum_{(i,j) \in \gamma} \left[i\rho_a \text{Tr}(\psi_i^\dagger \psi_j + \tau^x \psi_i^\dagger \sigma^x \psi_j + \tau^y \psi_i^\dagger \sigma^y \psi_j + \tau^z \psi_i^\dagger \sigma^z \psi_j) \right. \\
& + i\rho_c \text{Tr}(\psi_i^\dagger \psi_j + \tau^\gamma \psi_i^\dagger \sigma^\gamma \psi_j - \tau^\alpha \psi_i^\dagger \sigma^\alpha \psi_j - \tau^\beta \psi_i^\dagger \sigma^\beta \psi_j) + i\rho_d \text{Tr}(\tau^\alpha \psi_i^\dagger \sigma^\beta \psi_j + \tau^\beta \psi_i^\dagger \sigma^\alpha \psi_j) \\
& + i\rho_f \text{Tr}(\tau^\alpha \psi_i^\dagger \sigma^\gamma \psi_j + \tau^\gamma \psi_i^\dagger \sigma^\alpha \psi_j + \tau^\beta \psi_i^\dagger \sigma^\gamma \psi_j + \tau^\gamma \psi_i^\dagger \sigma^\beta \psi_j) + \text{H.c.} \Big] \\
& + \sum_{\langle\langle i,j \rangle\rangle} \left[t_3 \text{Tr}(\tau^z \psi_i^\dagger \psi_j) + \Delta_3 \text{Tr}(\tau^x \psi_i^\dagger \psi_j) + \text{H.c.} \right] + \sum_i \boldsymbol{\lambda}_i \cdot \text{Tr}(\psi_i \boldsymbol{\tau} \psi_i^\dagger)
\end{aligned} \tag{S1}$$

where ρ_a , ρ_c , ρ_d , and ρ_f come from the nearest neighbor interactions, t_3 and Δ_3 come from the J_3 Heisenberg exchange interactions, σ^m stands for the spin operation, τ^m denotes the gauge operation, and $\boldsymbol{\lambda}$ is imposed to ensure the SU(2) gauge invariance (the λ^z component is the Lagrangian multiplier for the particle number constraint).

Furthermore, to describe the long-range magnetic order, we introduce a background field $\mathbf{M}_i$ to enforce the symmetry-breaking. The ordering pattern is contained by the single- $\mathbf{Q}$ ansatz [10] with

$$\mathbf{M}_i = M \left(\sin \phi [\hat{\mathbf{e}}_x \cos(\mathbf{Q} \cdot \mathbf{r}_i) + \hat{\mathbf{e}}_y \sin(\mathbf{Q} \cdot \mathbf{r}_i)] + \cos \phi \hat{\mathbf{e}}_z \right),$$

where $\mathbf{Q}$ is the ordering momentum, $\hat{\mathbf{e}}_{x,y,z}$ are the local spin axes, and ϕ is the canting angle. The ordering momentum $\mathbf{Q}$ is adopted either from the classical ground state. For a given $\mathbf{Q}$, the local axes $\hat{\mathbf{e}}_{x,y,z}$ are fixed as they are in the classical state, while M and ϕ are treated as variational parameters.

Hence, the complete trial mean-field Hamiltonian for the K - J_1 - Γ - Γ' - J_3 model in an external magnetic field reads

$$H_{\text{mf}}^{\text{total}} = H_{\text{mf}}^{\text{QSL}} + \frac{1}{2} \sum_i (\mathbf{M}_i + \tilde{g} \mu_B \tilde{\mathbf{B}}) \cdot \text{Tr}(\psi_i^\dagger \frac{\boldsymbol{\sigma}}{2} \psi_i), \tag{S2}$$

where $\tilde{\mathbf{B}}$ is the effective Zeeman field due to the external magnetic field $\mathbf{B}$, but $\tilde{\mathbf{B}}$ is not necessarily equal to $\mathbf{B}$.

Then we perform Gutzwiller projection to the mean-field ground state $|\Psi_{\text{mf}}(\mathbf{R})\rangle$ to enforce the particle number constraint. The projected states $|\Psi(\mathbf{R})\rangle = P_G |\Psi_{\text{mf}}(\mathbf{R})\rangle$ provide a series of trial wave functions depending on the choice of the mean-field Hamiltonian $H_{\text{mf}}(\mathbf{R})$, where P_G denotes a Gutzwiller projection and $\mathbf{R}$ are treated as variational parameters. The energy of the trial state $E(\mathbf{R}) = \langle \Psi(\mathbf{R}) | H | \Psi(\mathbf{R}) \rangle / \langle \Psi(\mathbf{R}) | \Psi(\mathbf{R}) \rangle$ is computed using Monte Carlo sampling, and the optimal parameters $\mathbf{R}$ are determined by minimizing the energy $E(\mathbf{R})$.

In the variational process, we first choose a classical metastable configuration with $\mathbf{Q}$, then optimize the energy of the projected state. By comparing the optimal energies of different trial $\mathbf{Q}$ s, we obtain the approximate ground state. When the variational parameter $\mathbf{M}_i$ is zero, the magnetic order disappear, and system goes into QSL phase.

1. Magnitude dependence of the magnetic field

The phase diagram of the K - J_1 - Γ - Γ' - J_3 model with $K = 1.28\text{meV}$, $J_1/|K| = -1.09$, $\Gamma/|K| = -0.94$, $\Gamma'/|K| = 0.63$, $J_3/|K| = 0.94$ under applied in-plane magnetic field applied along the $\mathbf{a}^* = \frac{1}{\sqrt{2}}(\hat{x} - \hat{y})$ -direction is shown in Fig. S8.

Remarks: The adopted interaction parameters in this work are proportional to the $tx+$ set of parameter in Ref.[11], namely, $J_1 = -3.5\text{meV}$, $K = 3.2\text{meV}$, $\Gamma = -3.0\text{meV}$, $\Gamma' = 2\text{meV}$, excepted that the J_3 is enlarged to 3meV to ensure that the spin wave dispersion in the disordered phase is of concave shape at the Γ point (see section S6). Furthermore, to compare the excitation spectrum (see section S5B) with experiment, we then divide all the parameters by a factor 2.5. Comparing the VMC result with the phase diagram of Fig.1(a) in the main text, the theoretically obtained critical magnetic fields in Fig.S8, namely $B_{C1} = 0.47|K|/\mu_B \approx 10.47\text{T}$ and $B_{C3} = 0.77|K|/\mu_B \approx 17\text{T}$ are slightly larger than the experimental values. This is possibly caused by the simplified K - J_1 - Γ - Γ' - J_3 effective model where only two-body

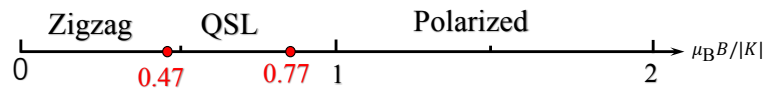


FIG. S8: **Calculated phase diagram with varying field strength.** The field is applied along the $\mathbf{a}^* = \frac{1}{\sqrt{2}}(\hat{x} - \hat{y})$ -direction, and the in-plane Lande g factor is adopted as $g = 4.13$ [5].

interactions are considered. The ignored multi-spin interactions may suppress the zigzag order and reduce the critical magnetic field. The region between B_{C1} and B_{C2} in the experiment is interpreted as a phase separation of zigzag phase and QSL phase, so in the VMC phase diagram it is replaced by a single phase transition.

Noticing that the partially polarized QSL phase and the polarized trivial phase have the same symmetry. The difference is that the QSL phase is Z_2 deconfined such that the spions and gauge fluxes are elementary excitations and the polarized trivial phase is Z_2 confined such that the elementary excitations are magnons. The deconfinement of the Z_2 gauge field can also be reflected from the degeneracy of the ground states on a torus. If the Z_2 gauge field is deconfined, then Z_2 fluxes are allowed low energy excitations. Consequently, there will be four-fold degenerate ground states on a torus (given that the Chern number of the mean field ground state is zero). On the other hand, if the Z_2 gauge field is confined, then there will be a single ground state on a torus.

The four-fold degeneracy in the Z_2 deconfined QSL phase can be understood as the following. Notice that the interactions are short-ranged, the energy density is a local quantity, but the global fluxes in the two holes are non-local. Hence in the thermodynamic limit the inserting of π -flux in one of the holes does not change the energy density of the system. The existence or absence of Z_2 fluxes through the two holes results in four-fold degenerate ground states on the torus.

Since the inserting of a π flux in one of the holes is equivalent to change the boundary condition of the mean field Hamiltonian in the corresponding direction from the periodic one to anti-periodic one (vice versa). If we note periodic and anti-periodic boundary conditions as $+$ and $-$ respectively, then there are four different combinations of boundary conditions for the x - and y - directions, namely $(+, +)$, $(+, -)$, $(-, +)$, $(-, -)$. The four boundary conditions results in four projected states, namely $|\psi_\alpha\rangle = P_G|\alpha\rangle_{\text{mf}}$ where $|\alpha\rangle_{\text{mf}}$ denotes the mean field ground state with boundary condition α . If the ground state degeneracy (GSD) of the spin model on the torus is four, then $|\psi_{(+,+)}\rangle, |\psi_{(+,-)}\rangle, |\psi_{(-,+)}\rangle, |\psi_{(-,-)}\rangle$ should be linearly independent. Otherwise, if the GSD is 1, then the above four states are essentially the same.

To judge the GSD, we calculate the fidelity matrix of the above four states with $\rho_{\alpha\beta} = \langle\psi_\alpha|\psi_\beta\rangle$. The four eigenvalues of the matrix ρ reflects the GSD. If all of the eigenvalues are of order 1, as shown in Fig. S9(a) for the partially polarized QSL phase (the fact that last eigenvalue is quite small may owe to the small Z_2 -flux excitation gap and the finite size effect), then the four states are linearly independent and consequently the GSD is four. The four-fold degeneracy of the ground states on a torus indicates the deconfinement of the Z_2 gauge field. On the other hand, if one of the eigenvalue is close to 4 and the other three are close to 0, as shown in Fig. S9(b) for the polarized trivial phase, then the GSD is one which indicates the Z_2 confinement.

2. Angular dependence of the magnetic field

Now we fix the strength of the external magnetic field $\mathbf{B}$ and vary its direction. Then we study the off-diagonal magnetization of the ground state, namely the magnetization in the direction perpendicular to $\mathbf{B}$. The off-diagonal

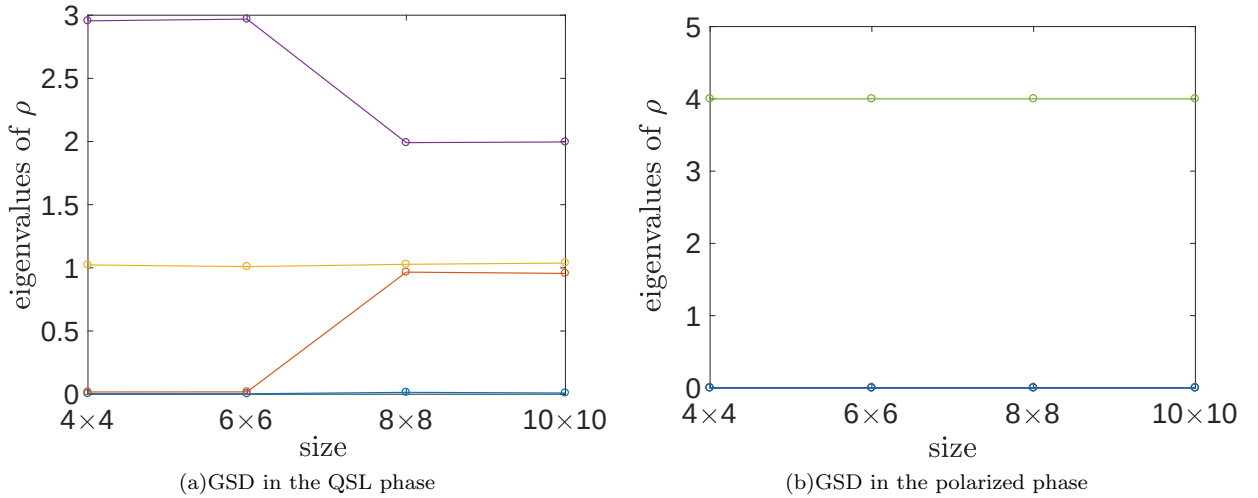


FIG. S9: **Eigenvalues of the fidelity matrix for the ground states on a torus.** (a) In the QSL phase, all the eigenvalues are approaching 1 with the increasing size, indicating that in thermodynamic limit the GSD is 4 and the Z_2 gauge field is deconfined. (b) In the polarized phase, the GSD is one, indicating Z_2 confinement.

magnetization, which can be read out from the variational parameters $\mathbf{M}_\perp \propto \tilde{\mathbf{B}}_\perp = \tilde{\mathbf{B}} \times \mathbf{B}/|\mathbf{B}|$, is closely related to the experimentally measured torque $\boldsymbol{\tau} = \mu_B \mathbf{M} \times \mathbf{B} = \mu_B \mathbf{M}_\perp \times \mathbf{B}$.

In the partially polarized QSL phase, there is no symmetry breaking. Therefore, when changing the angle between $\mathbf{B}$ and $\mathbf{a}^*$, it is expected that the angle dependence of the off-diagonal magnetization should respect all the symmetries of the Honeycomb lattice. The numerical result is shown in Fig. S10, which indicates a 6-fold periodicity and is consistent with the expectation.

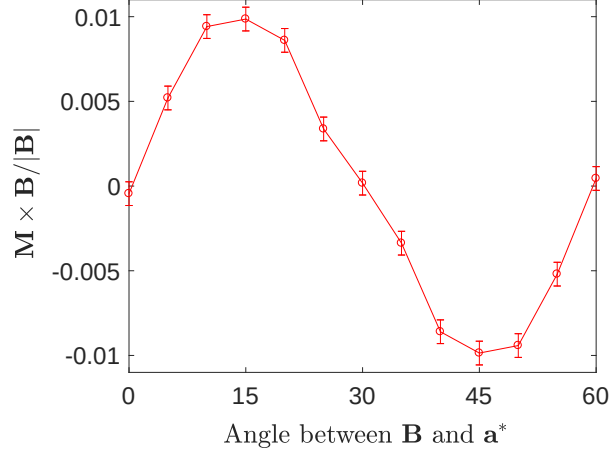


FIG. S10: **Calculated angle-dependent off-diagonal magnetization with fixed field strength.** The angle-dependence exhibits a 6-fold periodicity.

B. Excited states and Dynamical structure factor

1. Mean-field spectrum of the DSF

From fluctuation-dissipation relation, the dynamical structure factor (DSF) $S^{mn}(\mathbf{k}, \omega)$, $m, n = x, y, z$ is defined by

$$S^{mn}(\mathbf{k}, \omega) = -\frac{1}{\pi} [1 + n_B(\omega)] \chi''^{mn}(\mathbf{k}, \omega),$$

where $n_B(\omega)$ is the Bose-Einstein distribution and $\chi''^{mn}(\mathbf{k}, \omega) = \lim_{\delta \rightarrow 0} \text{Im } \chi^{mn}(\mathbf{k}, \omega + i\delta)$ is the imaginary part of $\chi^{mn}(\mathbf{k}, \omega)$ which is the analytic continuation of the finite temperature susceptibility $\chi^{mn}(\mathbf{k}, i\omega)$. As an example, we illustrate the calculation of $\chi^{+-}(\mathbf{k}, i\omega)$ in the following

$$\begin{aligned} \chi^{+-}(\mathbf{k}, i\omega) &= \int_0^\beta \langle T_\tau S_{\mathbf{k}}^+(\tau) S_{-\mathbf{k}}^-(0) \rangle e^{i\omega\tau} d\tau \\ &= \int_0^\beta \sum_{\mathbf{q}, \alpha, \beta} \langle T_\tau c_{\uparrow, \mathbf{q}}^{\alpha\dagger}(\tau) c_{\downarrow, \mathbf{q}+\mathbf{k}}^\alpha(\tau) c_{\downarrow, \mathbf{p}+\mathbf{k}}^{\beta\dagger}(0) c_{\uparrow, \mathbf{p}}^\beta(0) \rangle e^{i\omega\tau} d\tau \end{aligned}$$

where $S^\pm = S_x \pm iS_y$ and the superscript α, β sums over the A, B sub-lattices, T_τ means time order and $\langle \rangle$ stands for thermal average. If we abbreviate the spin, sub-lattice and particle-hole indices of C -fermions (here $C_{\mathbf{k}} = (c_{\uparrow A, \mathbf{k}}, c_{\downarrow A, \mathbf{k}}, c_{\uparrow B, \mathbf{k}}, c_{\downarrow B, \mathbf{k}}, c_{\uparrow A, -\mathbf{k}}, c_{\downarrow A, -\mathbf{k}}, c_{\uparrow B, -\mathbf{k}}, c_{\downarrow B, -\mathbf{k}})^T$) in $\chi^{+-}(\mathbf{k}, i\omega)$ as a, b, c, d , namely,

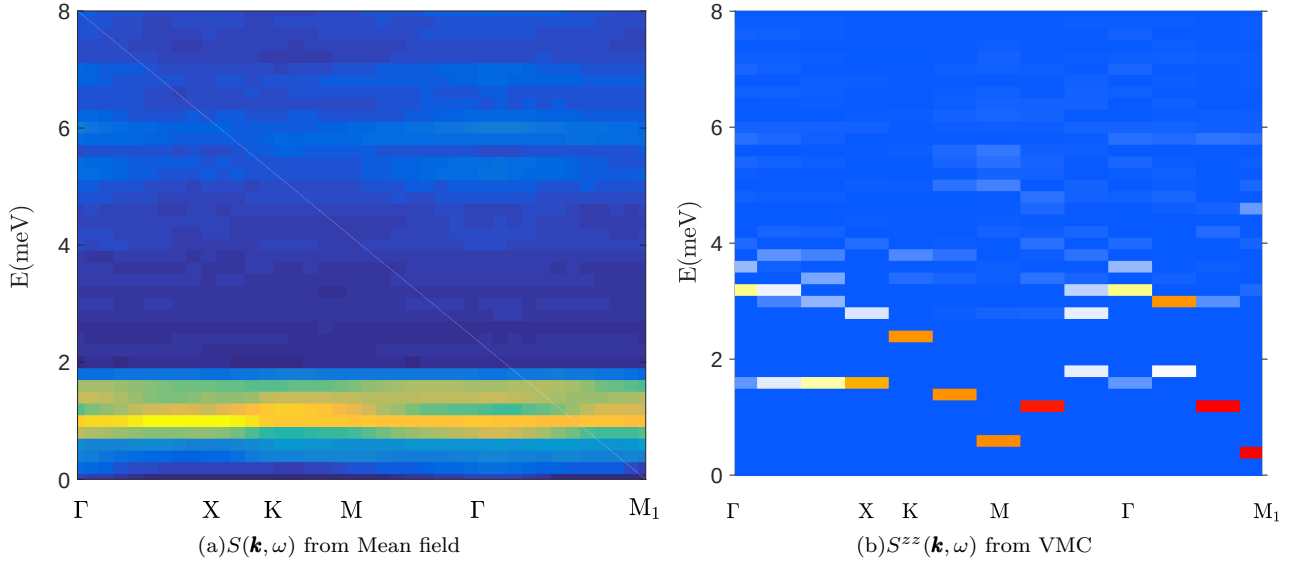


FIG. S11: **Calculated dynamic structure factors.** The dynamic structure factors calculated from (a) mean field states with $30 \times 30 \times 2$ sites and (b) Gutzwiller projected states with $6 \times 6 \times 2$ sites, respectively.

$\langle T_\tau c_{\uparrow, \mathbf{q}}^{\alpha \dagger}(\tau) c_{\downarrow, \mathbf{q}+\mathbf{k}}^\alpha(\tau) c_{\downarrow, \mathbf{p}+\mathbf{k}}^{\beta \dagger}(0) c_{\uparrow, \mathbf{p}}^\beta(0) \rangle = \langle T_\tau c_{\mathbf{q}}^{a \dagger}(\tau) c_{\mathbf{q}+\mathbf{k}}^b(\tau) c_{\mathbf{p}+\mathbf{k}}^{c \dagger}(0) c_{\mathbf{p}}^d(0) \rangle$, using Wick theorem, we have,

$$\begin{aligned}
 \chi^{+-}(\mathbf{k}, i\omega) &= \frac{1}{V} \int_0^\beta \sum_{\mathbf{q}, \{abcd\}} \langle T_\tau c_{\mathbf{q}}^{a \dagger}(\tau) c_{\mathbf{q}+\mathbf{k}}^b(\tau) c_{\mathbf{p}+\mathbf{k}}^{c \dagger}(0) c_{\mathbf{p}}^d(0) \rangle e^{i\omega\tau} d\tau \\
 &= \frac{1}{V} \int_0^\beta \sum_{\mathbf{q}, \{abcd\}} \left[-\langle T_\tau c_{\mathbf{p}}^d(0) c_{\mathbf{q}}^{a \dagger}(\tau) \rangle \langle T_\tau c_{\mathbf{q}+\mathbf{k}}^b(\tau) c_{\mathbf{p}+\mathbf{k}}^{c \dagger}(0) \rangle \right. \\
 &\quad \left. + \langle T_\tau c_{\mathbf{p}+\mathbf{k}}^{c+4}(0) c_{\mathbf{q}}^{a \dagger}(\tau) \rangle \langle T_\tau c_{\mathbf{q}+\mathbf{k}}^b(\tau) c_{\mathbf{p}}^{d+4, \dagger}(0) \rangle \right] e^{i\omega\tau} d\tau \\
 &= \frac{1}{V} \int_0^\beta \sum_{\mathbf{q}, \{abcd\}} \left[-\langle T_\tau c_{\mathbf{q}}^d(0) c_{\mathbf{q}}^{a \dagger}(\tau) \rangle \langle T_\tau c_{\mathbf{q}+\mathbf{k}}^b(\tau) c_{\mathbf{q}+\mathbf{k}}^{c \dagger}(0) \rangle \right. \\
 &\quad \left. + \langle T_\tau c_{-\mathbf{q}}^{c+4}(0) c_{\mathbf{q}}^{a \dagger}(\tau) \rangle \langle T_\tau c_{\mathbf{q}+\mathbf{k}}^b(\tau) c_{-(\mathbf{q}+\mathbf{k})}^{d+4, \dagger}(0) \rangle \right] e^{i\omega\tau} d\tau
 \end{aligned}$$

At mean field lever, the C fermions are diagonalized into the Bogoliubov particles, and the dynamic structure factors can be evaluated numerically. The complete dynamic structure factor $S(\mathbf{k}, \omega) = \frac{1}{2}(S^{+-}(\mathbf{k}, \omega) + S^{-+}(\mathbf{k}, \omega)) + S^{zz}(\mathbf{k}, \omega)$ for a system with $30 \times 30 \times 2 = 1800$ sites is shown in Fig.11(a), where the spinon continuum can be clearly seen in the range $\omega < 3\text{meV}$.

2. VMC results of the DSF

Now we try to construct the low-energy excitations via Gutzwiller projected spinon excited states

$$|\mathbf{k}; (\mathbf{k}-\mathbf{q})_m, (\mathbf{q})_n\rangle = P_G f_{\mathbf{k}-\mathbf{q}, m}^\dagger f_{\mathbf{q}, n}^\dagger |\Psi_{\text{mf}}\rangle, \quad (\text{S3})$$

where $f_{\mathbf{k}-\mathbf{q}, m}$ and $f_{\mathbf{q}, n}$ are eigen particles of the mean-field Hamiltonian. Here m and n are the band indices, $\mathbf{k}$ and $\mathbf{q}$ are the lattice momentum according to the translation operators.

The above two-spinon excitations form a continuum and do not correctly describe the low-energy excitations of the system. The failure of the state (S3) in describing the low-energy excitations is owing to the strong gauge interactions between the spinons which are not correctly addressed. To partially solve this problem, we diagonalize the original Hamiltonian in the subspace spanned by the two-spinon excitation continuum. Specifically, we calculate the matrix elements of the matrix $\mathcal{H}(\mathbf{k})$ with

$$\mathcal{H}_{pmn, qm'n'}(\mathbf{k}) = \langle \mathbf{k}; (\mathbf{k}-\mathbf{p})_m, (\mathbf{p})_n | H | \mathbf{k}; (\mathbf{k}-\mathbf{q})_{m'}, (\mathbf{q})_{n'} \rangle,$$

where $\mathbf{k}$ is the total momentum of the state with a pair of spinon excitations.

Since the states in the two-spinon continuum are not orthogonal, we need to calculate the metric matrix g formed by the overlap of the states,

$$g_{pmn, qm'n'}(\mathbf{k}) = \langle \mathbf{k}; (\mathbf{k} - \mathbf{p})_m, (\mathbf{p})_n | \mathbf{k}; (\mathbf{k} - \mathbf{q})_{m'}, (\mathbf{q})_{n'} \rangle.$$

Hence, the eigenproblem of $\mathcal{H}(\mathbf{k})$ should be calculated by

$$g^{-1}(\mathbf{k})\mathcal{H}(\mathbf{k})U = U \cdot \text{diag}(\epsilon_1, \epsilon_2, \dots),$$

where the eigenvalues $\epsilon_1, \epsilon_2, \dots$ are the ‘renormalized’ energy of the excitations.

The renormalized eigenfunction $|\mathbf{k}\rangle_{\text{rn}}$ is given by

$$|\mathbf{k}\rangle_{\text{rn}} = \sum_{\mathbf{q} \in BZ} \mathcal{F}(qmn) |\mathbf{k}; (\mathbf{k} - \mathbf{q})_m, (\mathbf{q})_n\rangle,$$

where $\mathcal{F}(qmn)$ is the eigenvector of the matrix $\mathcal{H}(\mathbf{k})$.

The more reliable DSF can be calculated using the renormalized energy and eigenstates. For instance, the zz -component of the DSF is expressed as

$$S^{zz}(\mathbf{q}, \omega) = \sum_n |\langle \Psi_n^{\mathbf{q}} | S_{\mathbf{q}}^z | \Psi_0 \rangle|^2 \delta(\omega - E_n^{\mathbf{q}} + E_0)$$

where $|\Psi_0\rangle$ is the variational ground state with energy E_0 and $|\Psi_n^{\mathbf{q}}\rangle$ is the n -th ‘renormalized’ two-spinon excited state with momentum $\mathbf{q}$ and energy $E_n^{\mathbf{q}}$. We note that $S_{\mathbf{q}}^z = \frac{1}{\sqrt{L}} \sum_{\mathbf{r}} \exp[i\mathbf{q} \cdot \mathbf{r}] S_{\mathbf{r}}^z$ is the Fourier-transformed spin operator for the component z , with L being the number of sites in the system.

However, due to the ‘renormalizing’ process, the computational complexity increases dramatically with system size. The data shown in Fig.11(b) is the result for a system with size $6 \times 6 \times 2 = 72$ sites. In Fig.11(b), the magnon modes as bound states of spinons can be clearly seen. However, due to finite size effect, the excitation gap of the magnons and the energy of the spinon continuum are exaggerated.

Noticing that the spinons are deconfined in the low-energy limit, the mean field results are qualitatively correct (but the bound states cannot be obtained at mean field level). Therefore, in the main text, we have combined the mean field result (with the spinon continuum) and the VMC result (with the magnon modes) to estimate the dynamic structure factor at large size limit.

S6. LINEAR SPIN WAVE

A. Ordered phase

In the zigzag phase, the magnetic unit cell contains 4 sites, as shown in Fig.S12(a). To obtain the linear spin wave (LSW) dispersion, we firstly adopt a new spin frame such that the new z -axis are parallel to the zigzag order, namely

$$\begin{pmatrix} S_i'^x \\ S_i'^y \\ S_i'^z \end{pmatrix} = \mathcal{R}_\alpha \begin{pmatrix} S_i^x \\ S_i^y \\ S_i^z \end{pmatrix} \quad (\text{S4})$$

where $\mathcal{R}_\alpha$ is a $SO(3)$ matrix with $\alpha = \text{R, B}$ stands for the red/blue sublattice index. Notice that the new z -axes in the red sublattice and the blue sublattice are opposite to each other. Thus the original K - J_1 - Γ - Γ' - J_3 model is transformed into a new one under the above rotations,

$$H = \sum_{\langle i, j \rangle} \mathbf{S}_i'^T H'^{ij} \mathbf{S}_j' - g\mu_B \sum_i \mathbf{B}_i' \cdot \mathbf{S}_i', \quad (\text{S5})$$

where $H'^{ij} = \mathcal{R}_i H^{ij} \mathcal{R}_j^T$ (with H^{ij} the original interaction matrix on the ij -bond), $\mathbf{B}_i' = \mathcal{R}_i \mathbf{B}$ with $\mathcal{R}_i = \mathcal{R}_R$ if i belong to the red sublattice and $\mathcal{R}_i = \mathcal{R}_B$ for the blue sublattice, and $g = 4.13$ [5].

In the new spin frame, we adopt the Holstein–Primakoff transformation $S_i'^x \sim \frac{1}{2}(a_i^\dagger + a_i)$, $S_i'^y \sim \frac{i}{2}(a_i^\dagger - a_i)$, $S_i'^z = \frac{1}{2} - a_i^\dagger a_i$ on the A-sublattice and $S_i'^x \sim \frac{1}{2}(b_i^\dagger + b_i)$, $S_i'^y \sim \frac{i}{2}(b_i^\dagger - b_i)$, $S_i'^z = \frac{1}{2} - b_i^\dagger b_i$ on the B-sublattice, where the bosons satisfy the usual commutation relations $[a_i, a_j^\dagger] = \delta_{ij}$, $[b_i, b_j^\dagger] = \delta_{ij}$ and $[a_i, b_j^\dagger] = 0$. Substituting the above

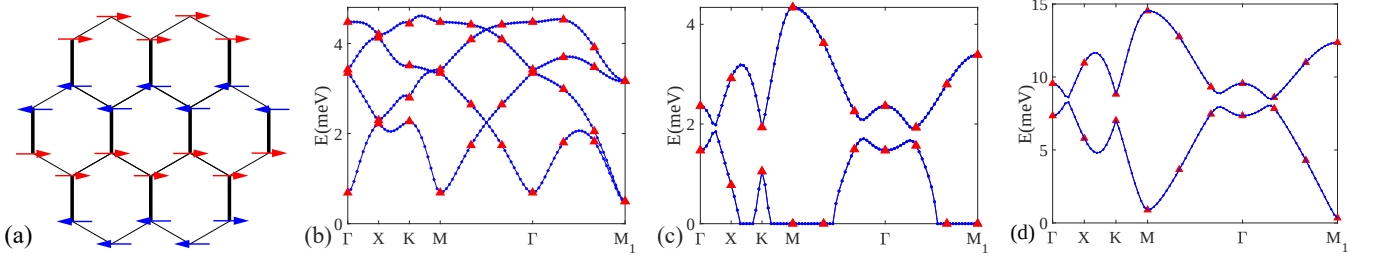


FIG. S12: **Linear spin wave dispersion.** (a) The Zigzag order on honeycomb lattice. (b) Linear spin wave of the Zigzag phase at zero field. (c) Linear spin wave of the disordered phase at $\mu_B B / |K| = 0.56$. (d) Linear spin wave of the polarized phase at $\mu_B B / |K| = 0.95$.

formulas into the rotated Hamiltonian (S5) and keeping the quadratic terms, we obtain the following Hamiltonian on the Fourier bases,

$$H_{\text{SW}} = \sum_{\mathbf{k}} \Psi^\dagger(\mathbf{k}) \mathcal{H}(\mathbf{k}) \Psi(\mathbf{k}), \quad (\text{S6})$$

where $\Psi^\dagger(\mathbf{k}) = (a_{R,\mathbf{k}}^\dagger, b_{B,\mathbf{k}}^\dagger, a_{B,\mathbf{k}}^\dagger, b_{R,\mathbf{k}}^\dagger, a_{R,-\mathbf{k}}, b_{B,-\mathbf{k}}, a_{B,-\mathbf{k}}, b_{R,-\mathbf{k}})$ with $a_{R/B,\mathbf{k}}^\dagger$ ($b_{R/B,\mathbf{k}}^\dagger$) being the magnon creation operator on A (B) and red/blue sublattices. Diagonalizing the above Hamiltonian using the Bosonic Bogoliubov transformation, we obtain the magnon excitations spectrum at zero field, as shown in Fig.S12(b).

B. Disordered phases

In the disordered phases, the spins are (partially) polarized due to the magnetic field. One can also calculate the spin wave spectrum assuming that the fully polarized state is the classical ground state. To this end, we perform a uniform orthogonal transformation to rotate the new z -axis to the field direction ($\mathbf{a}^*$), and

$$\begin{pmatrix} S_i^x \\ S_i^y \\ S_i^z \end{pmatrix} = \begin{pmatrix} \frac{1}{\sqrt{6}} & \frac{1}{\sqrt{6}} & \frac{-2}{\sqrt{6}} \\ \frac{1}{\sqrt{3}} & \frac{1}{\sqrt{3}} & \frac{1}{\sqrt{3}} \\ \frac{1}{\sqrt{2}} & \frac{-1}{\sqrt{2}} & 0 \end{pmatrix} \begin{pmatrix} S_i^x \\ S_i^y \\ S_i^z \end{pmatrix}, \quad (\text{S7})$$

and then introduce the Holstein–Primakoff transformation to obtain the spin wave Hamiltonian and the corresponding spectrum.

In the partially polarized QSL region (for instance at $\mu_B B / |K| = 0.56$), the LSW based on the fully polarized state is unstable, namely, the magnon spectrum has nonzero imaginary part when the real part goes to zero, as shown in Fig.S12(c). The instability of the spin wave indicates that the polarized state is not the ground state in the intermediate field region. In contrast, when the field is strong enough (for instance at $\mu_B B / |K| = 0.95$), the LSW based on the polarized state is fully gapped and stable [see Fig.S12(d)].

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