Supplementary information for Fractional and composite excitations of antiferromagnetic quantum spin trimer chains

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Supplementary Note 1: Exact solutions of small systems

In addition to the QMC-SAC calculations, we have also applied the ED method to obtain reliable results. ED is the most basic numerical method for determining the eigenvalues and eigenstates of a quantum magnet with up to tens of spin-1/2 spins. If all the eigenstates have been obtained for a very small system, Eq. (3) in main text can be computed directly, and somewhat larger systems can be considered within the Lanczos ground-state method combined with the continued fraction expansion [1]. To reach larger sizes, we can further use symmetries to block diagonalize the Hamiltonian and reduce the computation time and memory requirement. Although ED is limited to extracting exact information from relatively small systems, where finite-size effects are still significant, we can gain more reliable insights pertaining to the thermodynamic limit by comparing results from the two different approaches.

In Supplementary Figure 1, we present the spin excitation spectra of the trimer chain obtained by ED calculations for different values of the coupling ratio g. Here the ED results are calculated from a chain with 30 spins. We have applied broadening in the momentum direction to aid in the visualization of spectrum.

In the ED calculations we can clearly see fine structure due to the small number of δ-functions (which are broadened for visualization) in Eq. (3) of main text. The visible bands formed by the individual excitations of the small chain fall within the spinon continuum forming in the thermodynamic limit. The ED and QMC-SAC spectral weight distributions are similar, but the QMC-SAC exhibit the full spinon continua more clearly. The two distinct excitation branches above the spinon continuum are very similar in the two sets of results.

Supplementary Note 2: Effective Heisenberg coupling

To derive the effective HAC arising from the doublet trimer ground states, we formally split the original Hamiltonian into three-spin blocks and apply the generic Kadanoff method [2–5]. The trimer chain Hamiltonian is thus decomposed into the intratrimer (H^{t}) and intertrimer (H^{tt}) parts. The lowest eigenstates of each trimer are then used to construct the basis for the effective Hilbert space. To achieve an effective Hamiltonian with structure similarity to the original one, the full Hamiltonian is projected onto the effective Hilbert space. The intratrimer Hamiltonian, which only contains the J_1 couplings, reads

$$H^{\mathrm{t}} = J_1 \sum_{j=1}^{N} \left(\mathbf{S}_{j,a} \cdot \mathbf{S}_{j,b} + \mathbf{S}_{j,b} \cdot \mathbf{S}_{j,c} \right), \tag{1}$$

where N represents the number of trimers and we again refer to Fig. 1 of main text for the definition of the intratrimer labels a, b, c. The intertrimer Hamiltonian is given by

$$H^{\mathrm{tt}} = J_2 \sum_{j=1}^{N-1} \mathbf{S}_{j,c} \cdot \mathbf{S}_{j+1,a}, \tag{2}$$

which only contains the interaction between the c site of jth trimer and the a site of $(j+1)$th trimer. Each trimer has two degenerate ground states (see Fig. 3 in main text);

$$|0\rangle^1 = \frac{1}{\sqrt{6}} \left(|\uparrow\downarrow\downarrow\rangle - 2\,|\downarrow\uparrow\downarrow\rangle + |\downarrow\downarrow\uparrow\rangle \right), \tag{3}$$

$$|0\rangle^2 = \frac{1}{\sqrt{6}} \left(|\downarrow\uparrow\uparrow\rangle - 2\,|\uparrow\downarrow\uparrow\rangle + |\uparrow\uparrow\downarrow\rangle \right), \tag{4}$$

where $|\uparrow\rangle$ and $|\downarrow\rangle$ are the eigenstates of the spin operator σ_z. The projection operator of the jth trimer,

$$\mathbf{P}_j = |0\rangle_j^2 \langle\Uparrow|_j + |0\rangle_j^1 \langle\Downarrow|_j \tag{5}$$

is built to project the Hamiltonian onto the low-energy subspace, where $|\Uparrow\rangle$ and $|\Downarrow\rangle$ are the renamed base kets in this effective Hilbert space. The effective Hamiltonian up to the first-order correction is

$$H_{\mathrm{eff}} = \mathbf{P}^\dagger H^{\mathrm{t}} \mathbf{P} + \mathbf{P}^\dagger H^{\mathrm{tt}} \mathbf{P}, \tag{6}$$

where the total projection operator is $\mathbf{P} = \prod_{j=1}^{N} \mathbf{P}_j$. The effective Pauli operators are given by

$$\widetilde{\sigma}_j^\beta = \xi_i^\beta \mathbf{P}_j^\dagger \sigma_{i,j}^\beta \mathbf{P}_j, \quad (i = a, b, c;\ \beta = x, y, z), \tag{7}$$

where ξ_i^β is the coefficient required to preserve the SU(2) algebra. Inserting the effective Pauli operators into Eq. (6), we obtain the effective Hamiltonian

$$H_{\mathrm{eff}} = J_{\mathrm{eff}} \sum_{j=1}^{N} \widetilde{\mathbf{S}}_j \cdot \widetilde{\mathbf{S}}_{j+1}, \tag{8}$$

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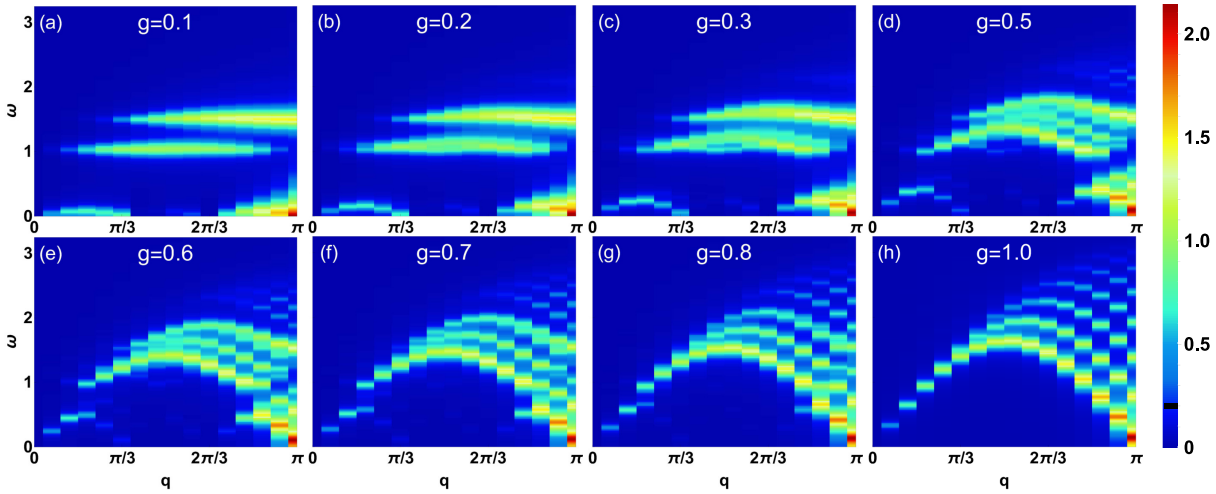


Supplementary Figure 1. Dynamic spin structure factor $\mathcal{S}(q,\omega)$ obtained by ED for the trimer chain with $L = 30$ spins ($N = 10$ unit cells) at different coupling ratios g. The boundary between the linear and logarithmic color mappings is $U_0 = 0.2$, as indicated on the color bar. In this ED calculation a Lorentzian broadening factor 0.05 was used of the δ functions in Eq. (3) of main text, and an uniform stretching of the 15 points in the momentum direction of the half BZ shown here was also applied by the software used to construct the graph.

which describes an isotropic HAC in which the effective coupling strength only depends on the intertrimer coupling, $J_{\text{eff}} = 4J_2/9$.

Supplementary Note 3: Dispersion relations

As we have discussed in the main text, the internal excitations of the trimer chain are almost localized when g is small, and cannot be classified as magnons or triplons. In order to obtain further insight into the nature of these excitations, we propose a scheme to calculate their dispersion relations. The results are shown in Fig. 4 of main text as a number of dispersion relations corresponding to propagation of internal trimer excitations in the background of a chain with defects mimicking the complex ground state of the effective HAC. The overall good agreement with the QMC–SAC results on the location of these excitations and their band widths suggest that our picture of the excitation is correct even though the calculation involves a very rough approximation of the ground state and a simple ansatz for its excitations.

We begin by assuming that the ground-state wave function of spin chain is a product state of ground states of each trimer,

$$\left|\psi_{\text{g}}\right\rangle = |0\rangle_1 \, |0\rangle_2 \cdots |0\rangle_r \cdots |0\rangle_N \,, \tag{9}$$

where $|0\rangle$ is one of the two ground states of a single trimer. Thus, we neglect the interactions that cause the effective HAC with its spinon continuum, and instead deal with 2^N degenerate ground states without enforcing any quantum numbers (spin or momentum). Note that we do not apply degenerate perturbation theory here, because we are targeting excitations above the 2^N-fold degenerate ground-state manifold. We also do not carry out a formal perturbation expansion, but construct intuitive variational states including the internal excitations of one trimer.

If the rth trimer is excited from its ground state $|0\rangle$ to one of its higher states $|1\rangle$, then the wave function with this localized excitation reads

$$|\psi_{\text{e}}\rangle_r = |0\rangle_1 \, |0\rangle_2 \cdots |1\rangle_r \cdots |0\rangle_N \,. \tag{10}$$

We can give this excitation a momentum and calculate its dispersion relation. Since the trimer has two types of excitations (see Fig. 3 of main text), we will obtain two bands. Next, we first consider the lower $\omega = J_1$ doublon excitation of the isolated trimer, and then defer the similar but more complicated case of the $\omega = 3J_1/2$ quarton excitation.

A. Intermediate-energy doublon excitation

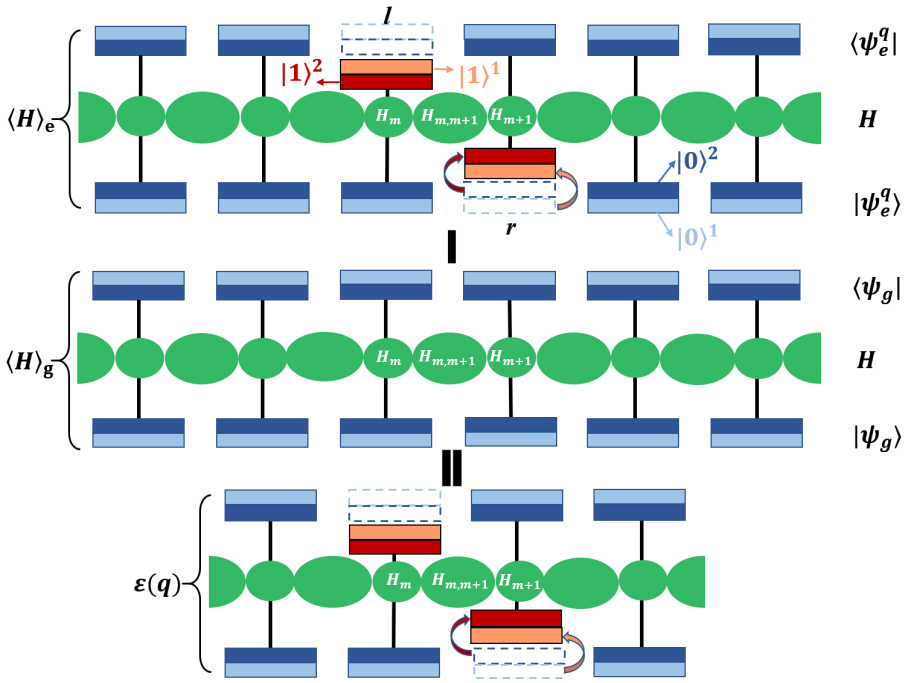
For simplicity, we first consider the approximate uniform ground-state wave function with all the trimers in the same ground state of the isolated trimer, $|0\rangle^2 = (|{\downarrow\uparrow\uparrow}\rangle - 2\,|{\uparrow\downarrow\uparrow}\rangle + |{\uparrow\uparrow\downarrow}\rangle)/\sqrt{6}$. This state clearly has momentum $q = 0$. In the lowest excitation involving the internal trimer excitation, one of the trimers is put in its first excited state $|1\rangle^1 = (|{\downarrow\downarrow\uparrow}\rangle - |{\uparrow\downarrow\downarrow}\rangle)/\sqrt{2}$ with $\Delta M = -1$. Fourier transforming such a state, the one-trimer excited state in momentum space is given by

$$\left|\psi_{\text{e}}^{q}\right\rangle = \frac{1}{\sqrt{N}} \sum_{r=1}^{N} \text{e}^{-\text{i}qr} \, |\psi_{\text{e}}\rangle_{\text{r}} \,. \tag{11}$$

The total Hamiltonian with periodic boundary conditions is the summation of intratrimer Hamiltonian H_m and the intertrimer Hamiltonian $H_{m,m+1}$

$$H = \sum_{m=1}^{N} H_m + \sum_{m=1}^{N} H_{m,m+1}. \tag{12}$$

Next, we calculate the expectation values of this Hamiltonian in the ground state and the first excited trimer momentum



Supplementary Figure 2. Graphical representation of the calculation of the dispersion relation $\epsilon(q) = \langle H\rangle_{\rm e} - \langle H\rangle_{\rm g}$ of the intermediate-energy excitation within our approximation. The doubly-degenerate trimer eigenstates are shown as lighter and darker blue (ground states) and orange or red (first excited states). With these states, the intermediate-energy excitations with $|\Delta M| = 1$ originate from a trimer ground state $|0\rangle_r^1$ on the trimer located at r when excited to $|1\rangle_r^2$ or $|0\rangle_r^2$ excited to $|1\rangle_r^1$ (and in the corresponding bra states we use the site index l instead of r). The surrounding trimers, which can be in either of their doubly-degenerate ground states, are not affected. The excitations are given momentum q, and the matrix elements contributing to the dispersion relation are indicated. In addition to the case $r = l + 1$ shown, $r = l - 1$ contributes equally and $r = l$ gives a different contribution. No other relationships between r and l contribute.

state. For the ground state of trimer chain, the expectation value of H trivially evaluates to

$$\langle H\rangle_{\rm g} = \left\langle\psi_{\rm g}\right| H \left|\psi_{\rm g}\right\rangle = NE_0 + \frac{N}{9}J_2. \tag{13}$$

For the one-trimer excited state, we consider separately the two terms of the expectation value of H in momentum space: $\left\langle\psi_{\rm e}^q\right| \sum_{m=1}^N H_m \left|\psi_{\rm e}^q\right\rangle$ and $\left\langle\psi_{\rm e}^q\right| \sum_{m=1}^N H_{m,m+1} \left|\psi_{\rm e}^q\right\rangle$. The first term only contributes the gap $E_1 - E_0$. To evaluate the second term, we let $H_{m,m+1}$ act on the c spin of mth trimer and the a spin of the $(m + 1)$th trimer. The excited trimers labeled by l and r in the bra $\left\langle\psi_{\rm e}^q\right|$ and ket $\left|\psi_{\rm e}^q\right\rangle$ should take all the values of $[1, N]$. Practically, only the cases $l = r$ and $l = r \pm 1$ contribute, however, because $\left\langle\psi_{\rm e}^q\right|$ and $\left|\psi_{\rm e}^q\right\rangle$ are orthogonal for $|l - r| \geq 2$. Therefore, the expectation value of H for the one-trimer excited state is

$$\begin{aligned}
\langle H\rangle_{\rm e} &= \left\langle\psi_{\rm e}^q\right| H \left|\psi_{\rm e}^q\right\rangle \\
&= \frac{1}{N}\sum_{r=1}^{N}\sum_{l=1}^{N} {\rm e}^{-{\rm i}qr+{\rm i}ql} \left\langle 0^2\cdots 1_l^1\cdots 0^2\right| \sum_{\rm m=1}^{\rm N} {\rm H_m} \\
&\quad + \sum_{m=1}^{N} H_{m,m+1} \left|0^2\cdots 1_r^1\cdots 0^2\right\rangle \\
&= (N-1)E_0 + E_1 + J_2\left(\frac{N-2}{9} - \frac{1}{3}\cos q\right).
\end{aligned} \tag{14}$$

Thus, the dispersion relation is of what we refer to as the intermediate-energy excitation in reduced BZ is

$$\begin{aligned}
\epsilon(q) &= \langle H\rangle_{\rm e} - \langle H\rangle_{\rm g} \\
&= E_1 - E_0 - \frac{2}{9}J_2 - \frac{1}{3}J_2\cos q,
\end{aligned} \tag{15}$$

independently of the length of the spin chain. Unfolding this dispersion relation by replacing q with $3q$, we can obtain the dispersion relation in full BZ

$$\epsilon(q) = E_1 - E_0 - \frac{2}{9}J_2 - \frac{1}{3}J_2\cos(3q). \tag{16}$$

The first two terms describe the gap of the localized excited trimer, and the last two terms reveal the dynamical features dominated by the intertrimer interaction. By symmetry, for the excitation from ground state $|0\rangle^1$ to the other first-excited state $|1\rangle^2$ with $\Delta M = 1$, the same dispersion relation, Eq. (15), is also obtained in the reduced BZ.

To better mimic the true many-body ground state, we can consider random arrangements of the states $|0\rangle_i^1$ and $|0\rangle_i^2$ on each site. Exciting from all possible state in this degenerate manifold will lead to a band of excitations that approximate the true bands arising from combinations of an internal trimer excitation and spinons. We always start from momentum 0 in the ground state, and in the excited state we consider the same configuration of $|0\rangle_i^1$ and $|0\rangle_i^2$ but with one of these

trimer ground states at site r excited to the first trimer excitation $|1\rangle_r^1$ or $|1\rangle_r^2$ such that $|\Delta M| = 1$, and this state is given momentum q. Because of the form of the Hamiltonian, and examining the energy calculation as illustrated in Supplementary Figure 2, we can conclude that the dispersion relations are mainly dependent on the excited trimers and their neighbours. Translated to finite size, this means that the correct generic result is already obtained from a system with $N = 4$ trimers. Thus, we can generate 16 different dispersion relations for the intermediate-energy excitation, and because of symmetries there are only four different ones. In addition to the one already given in Eq. (15), the other three are the Eqs. (6)-(8) in main text. Although all possibilities have been considered above, the true ground state for $g > 0$ is a total-spin singlet satisfying $M = \sum_i M_i = 0$, and the numbers of $|0\rangle^1$ and $|0\rangle^2$ used when forming ground states should therefore be equal. After considering this restriction, the result of Eq. (15) is eliminated since its approximation corresponds to a maximal-M uniform ground state. Only 6 dispersion relations described by Eqs. (6)-(8) in main text are left for $N = 4$ trimers. Each equation corresponds to two degenerate dispersion relations. All three dispersion relations are graphed in Fig. 5(a) of main text for $g = 0.1$. Two of the above dispersion relations are independent of q since the excitations are localized. Only the $l = r$ terms in Supplementary Figure 2 contribute to these excitations, while the $l = r \pm 1$ terms lead to propagation of the internal trimer excitations. According to the characters of this excitation starting from the $g = 0$ limit, we refer to it as the doublon.

B. Dispersion relations of the high-energy excitation

Calculating the dispersion of the high-energy excitations evolving from the trimer quartet at $\omega = 3J_1/2$ is more complicated but we follow the same strategy as for the doublon. In the latter case, the intermediate-energy band is formed by reconfiguring the singlet bond in the original ground state of the trimer in Fig. 3 of main text at a cost of J_1. The higher-energy quarton band is formed out of trimers excited into their $S = 3/2$ highest state, which can be seen as the singlet of two spins excited into a triplet, at a cost of $3J_1/2$. This rough estimation of the excited energy matches very well the numerical ED and QMC-SAC results for small g values.

Again our calculation does not conserve the total spin of the collective many-body state, but we consider the excitations corresponding to $\Delta S = 1$ on one trimer. In this calculation, we not only need to prepare the ground-state wave functions, but also select the excited states from the quadruplet. Here, $N = 4$ trimers are also adequate to do the calculation at our level of linear-in-g approximation. With each of the lth and rth excited trimers having four possible states, at least 2^4 ground states and 4^2 higher-lying excited states are needed, and there are $2^4 \times 4^2 = 256$ cases in total. After implementing the restriction of the ground-state wave function having $M = 0$, 96 cases are left. As a result, the dispersion relations of the high-energy excitation in the reduced BZ fall into 33 different cases and are displayed in Eqs. (9)-(17) in the main text.

Supplementary References

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